PHYSICAL REVIEW A VOLUME 17, NUMBER 4 APRIL 1978

Geometry of density matrices. II. Reduced density matrices and N representability

John E. Harriman
Theoretical Chemistry Institute, University of Wisconsin-Madison, Madison, Wisconsin 53706
(Received 28 April 1977)

The reduction of density matrices defined with respect to a finite basis set is considered. A basis set can be introduced into the vector space of Hermitian matrices acting on functions of the coordinates of p particles for each value of p. An analogy with the construction of spin eigenfunctions is used to obtain a particular basis set called the reducing basis. Reduction of one of these basis elements is either one to one or maps into the origin. Any element in the preimage of a density matrix can thus be resolved into two components, one uniquely determined by the density matrix and the other arbitrary within a certin subset. A restatement of the N-representability problem is given, and two sufficient conditions and one necessary condition for N representability are given in terms of distances and norms.

I. INTRODUCTION

In the preceding paper[1] (hereafter referred to as I), a space whose elements are Hermitian matrices of a fixed, finite dimension was considered. A metric and a set of basis elements or coordinate axes were introduced into the space. It was shown that the basis can be chosen so that only one of the basis matrices has nonvanishing trace. Matrices of unit trace are then readily identified as those having a fixed value of this one particular component. The remaining condition on a density matrix, that it be positive, is not so readily dealt with, but some of the geometric consequences were considered in I. In the present paper, we will consider a sequence of such spaces, corresponding to matrices of operators acting on functions for different numbers of electrons. The mapping operation corresponding to reduction of a density matrix will be treated, and it will be shown that a particular choice of basis elements in each space greatly simplifies the interpretation of the consequences of reduction and investigation of the N-representability problem. Some of the results developed here are implicit in earlier work of Kummer.[2] The present point of view is somewhat different, however, and will hopefully make the results more readily accessible to quantum chemists and physicists. The introduction of the basis set is novel and has significant consequences.

In Sec. II, basis functions will be introduced and some well-known, essentially trivial results will be presented briefly to establish notation and provide a foundation for further development. In Sec. III a new space is introduced. It combines the spaces for different numbers of electrons and is related to them as Fock space is related to Hilbert spaces.[3] In fact a second-quantized notation could be used, but it appears to offer little advantage in the present context. The total space can be divided into orthogonal subspaces which remain disjoint under the reduction operation. A set of operators, one combination of which corresponds to the reduction operation, is introduced and the operators of the set are shown to satisfy angular momentum commutation relations. This suggests that new basis elements be introduced in each subspace, corresponding to angular momentum eigenfunctions. It is shown that such a choice of basis cleanly separates the unique and nonunique parts of any element in the preimage of any reduced density matrix. These results are applied to an analysis of some aspects of the N-representability problem in Sec. IV.

Treatment in subsequent papers will include a more complete analysis of the relationships between one- and two-electron reduced density matrices and their spin components as considered from the geometric point of view, the Hamiltonian and other operators, and energy considerations.

II. PRELIMINARIES

The concept of reduction, which is well known in connection with density matrices of fermion or boson systems, can readily be extended to other operators which are expressible as integral operators with kernels that are permutationally symmetric or antisymmetric functions of a set of (particle) coordinates. Let $N=(1,2,\ldots,n)$ be the (ordered) set of integers from 1 through n, where each integer refers to the space and spin coordinates of the particle labeled by that integer; let $N'=(1',\ldots,n')$ be a similar set in which each coordinate is distinguished by a prime (from the corresponding unprimed coordinate) and let $N=(P,Q)$, where $P=(1,\ldots,p)$ is the set of the first p integers, corresponding to the coordinates of the first p particles and $Q=(p+1,\ldots,p+q=n)$ is similarly defined for any $p<n$. The symbols

$\int dN'$ will be used to indicate integration over the full range of all variables in the set labeled by N', etc.

Then if $\hat{F}^{(n)}$ is an operator acting on n-particle functions such that

$$\hat{F}^{(n)}\Psi^{(n)}(N) = \int F^{(n)}(N,N')\,\Psi(N')\,dN' , \tag{1}$$

we can define a reduced operator $\hat{F}^{(p)}$ acting on p-particle functions as

$$\hat{F}^{(p)}\Psi^{(p)}(P) = \int F^{(p)}(P,P')\,\Psi^{(p)}(P')\,dP' , \tag{2a}$$

where

$$F^{(p)}(P,P') = \int F^{(n)}(P,Q;P',Q)\,dQ . \tag{2b}$$

A density operator or density matrix is clearly of this form. In the present series of papers we are considering only operators restricted to the space spanned by a finite orthonormal basis set $\{\Phi_K\}$. Such an operator is equivalent to a matrix

$$\hat{F}^{(n)} \leftrightarrow \underline{F}^{(n)}, \quad F^{(n)}_{JK} = \int \Phi^{(n)*}_J \hat{F}^{(n)} \Phi^{(n)}_K\, dN , \tag{3}$$

and its effect can be given in integral operator form with kernel

$$F^{(n)}(N,N') = \sum_{J,K} F^{(n)}_{JK}\,\Phi_J(N)\,\Phi^*_K(N') . \tag{4}$$

Any operator restricted to the finite-dimensional space can be so expressed.

In the notation thus far used, N, P, Q, etc., refer to sets of integers, and implicitly to sets of particle coordinates; indices J,K,L, label functions of the basis set and will correspond to sets of integers labeling spin orbitals; $\hat{F}^{(n)}$ is an operator acting on n-particle functions, $\underline{F}^{(n)}$ is the corresponding matrix, and $F^{(n)}(N,N')$ the corresponding integral operator kernel.

The simplest way to introduce a consistent set of basis functions into each of a series of spaces referring to different numbers of identical particles is to begin with a one-particle basis set and construct appropriately symmetrized products. Only fermions will be considered here, and they will be referred to as "electrons," although the results would apply equally well to other fermion systems.

Let $\{\phi_k,\ k=1,2,\ldots,r\}$ be a set of orthonormal spin orbitals. The basis set of p-electron functions is then taken to be

$$\{\Phi^{(p)}_K(P) = A_p\,\phi_{k_1}(1)\phi_{k_2}(2)\cdots\phi_{k_p}(p)\}$$

where A_p is the normalizing p-particle antisymmetrizer and $K=(k_1,k_2,\ldots,k_p)$ is an ordered set of indices. There are $R_p=\binom{r}{p}$ orthonormal functions Φ^p_K. The number of indices in the set K is determined by the superscript index on the function labeled by this set label. We will refer to sets, and use set-theoretic notation for inclusion, union, etc., but an ordering must be assumed if the phase of $\Phi^{(p)}_K$ is to be uniquely determined. The ordering assumed for the set K is $k_1<k_2<\cdots<k_p$ unless otherwise specified, and whenever a new set is formed by union or by addition of an index, etc., it is assumed that the new set is immediately ordered in an appropriate way.

We denote by $\mathcal{S}_p$ the space whose elements are $R_p\times R_p$ Hermitian matrices. As pointed out above, these matrices may be thought of as corresponding to operators (Hermitian in this case) restricted to the space spanned by the $\{\phi^p_K\}$ and expressible as in Eq. (2). The dimension of $\mathcal{S}_p$ is R_p^2, and we introduce a basis for it as in I:

$\underline{A}^K(p)$:

$$[A^K(p)]_{IJ} = \delta_{IJ}\delta_{JK} , \tag{5a}$$
$$A^K(P,P') = \Phi^{(p)}_K(P)\,\Phi^{(p)*}_K(P') ;$$

$\underline{B}^{KL}(p)$:

$$[B^{KL}(p)]_{IJ} = (1/\sqrt{2})(\delta_{IK}\delta_{JL}+\delta_{IL}\delta_{JK}) ,$$
$$B^{KL}(P,P') = (1/\sqrt{2})[\Phi^{(p)}_K(P)\,\Phi^{(p)*}_L(P') + \Phi^{(p)}_L(P)\,\Phi^{(p)*}_K(P')] , \tag{5b}$$
$$K<L$$

$\underline{C}^{KL}(p)$:

$$[C^{KL}(p)]_{IJ} = (i/\sqrt{2})(\delta_{IK}\delta_{JL}-\delta_{IL}\delta_{JK}) ,$$
$$C^{KL}(P,P') = (i/\sqrt{2})[\Phi^{(p)}_K(P)\,\Phi^{(p)*}_L(P') - \Phi^{(p)}_L(P)\,\Phi^{(p)*}_K(P')] , \tag{5c}$$
$$K<L .$$

The superscript labels on the basis matrices A,B,C are associated with functions, as in paper I. In addition to the ordering of indices within each set, we must define an ordering of the set indices $J,K,L,\ldots$. We can, for example, associate with K a binary number of r digits, where the jth digit is one if index j is included in K and zero otherwise, and order sets containing the same number of indices so that the corresponding binary numbers will be in increasing order. The notation $K<L$ means that the set K comes before the set L in the standard ordering. In the basis elements $\underline{B}^{KL}$ and $\underline{C}^{KL}$ it is assumed that $K<L$ in this sense. [Cf. Eq. (11) of I]. Subscript labels $[\ ]_{IJ}$ identify matrix elements, so they are also associated with functions. The Kronecker deltas are defined in the usual way, but in this case the indices I, J, K, and L are associated with sets of spin oribtal indices, so that $\delta_{JK}=1$ if the set $(j_1,\ldots,j_p)$ is the same as the set $(k_1,\ldots,k_p)$ and is zero otherwise.

Thus, for example, $\underline{A}^K(p)$ is a diagonal matrix [of dimension $\binom{r}{p}^2$] with a 1 in row and column K and zero's elsewhere. As was noted in I, the basis defined in Eqs. (5) is orthonormal and complete for $\mathcal{E}_p$. It will be convenient to include as special cases the one-dimensional spaces $\mathcal{E}_0$ with single basis element $\underline{A}(0)=1$, and $\mathcal{E}_r$, with basis element $\underline{A}(r)$ given by Eq. (5a) where $\Phi^{(r)}$ is the antisymmetrized product of all the spin orbitals.

The reduction operation defined in Eqs. (1)–(4) provides a mapping of $\mathcal{E}_n$ onto $\mathcal{E}_p$. Since the basis set in each space is complete, the effect of reduction on any element of $\mathcal{E}_n$ will be determined if we specify the effect of reduction on each basis element of $\mathcal{E}_n$ in terms of basis elements of $\mathcal{E}_p$. A notation for the reduction operation is introduced by[4]

$$\underline{F}^{(q)}=\hat{L}_p^q\underline{F}^{(p)} \tag{6}$$

for any q, not necessarily $n-p$, with $0\leq q<p$. Then

$$\hat{L}_p^q\underline{A}^K(p)=\binom{p}{q}^{-1}\sum_{\overline{K}\in K}\underline{A}^{\overline{K}}(q)\,. \tag{7}$$

Here $\overline{K}$ is a subset of q indices (again ordered $\overline{k}_1<\cdots<\overline{k}_q$) chosen from among the p indices in K. There are $\binom{p}{q}$ such subsets and thus $\binom{p}{q}$ terms in the sum. This result is well-known and is trivial except possibly for the constant factor. A factor of $(p-q)!$ is associated with different orderings of the indices labeling those coordinates with respect to which integration has been carried out, and the other parts of the binomial coefficient convert one antisymmetrizer into the other. Note that either $\underline{A}^K(p)$ or $\underline{A}^{\overline{K}}(q)$ has associated with it two functions, Φ, and thus two antisymmetrizers contribute constant factors.

Before we consider the reduction of $\underline{B}$'s and $\underline{C}$'s, it will be convenient to introduce some additional notation associated with pairs of p-particle indices. A pair of such indices K,L will have some indices in common, which we denote by com(K,L) and some indices which differ. We will use K/L to denote the set of indices in K which do not occur in L, and L/K to denote the set of indices in L which do not occur in K. The number of indices in the set K/L or L/K will be called the *difference* of the pair K,L and will be denoted by d. As initially defined, K and L are ordered sets. We can define a new ordering, called the pair ordering, in which K/L, L/K and com(K,L) are each ordered within themselves, but in K all the indices in K/L come before those in com(K,L) and similarly the indices in L/K come first in L. The index-set pair, with this pair ordering, will be denoted by $[KL]$. Since reordering indices in K or L may change the signs of Φ_K,Φ_L, it is clear that

$$\underline{B}^{[KL]}(p)=\pm\underline{B}^{KL}(p) \tag{8}$$

and similarly for $\underline{C}$. The plus sign is appropriate if the permutations required to convert K and L from fully ordered to pair ordered have the same parity, and the minus sign is appropriate when these parities are different. We now use the pair-ordered sets to define basis elements; the effect will be to change the signs of some components in the expansion of any element in $\mathcal{E}_p$, but this occurs in a known way.

The effect of reduction on $\underline{B}^{[KL]}$ can then be expressed as

$$\begin{aligned}
&\hat{L}_p^q\underline{B}^{[KL]}(p)=0 \quad \text{if } d>q\,,\\
&\hat{L}_p^q\underline{B}^{[KL]}(p)=\binom{p}{q}^{-1}\underline{B}^{K/L,L/K} \quad \text{if } d=q\,,\\
&\hat{L}_p^q\underline{B}^{[KL]}(p)=\binom{p}{q}^{-1}\sum_M\underline{B}^{[IJ]} \quad \text{if } d<q\,,
\end{aligned} \tag{9}$$

where $M\subset\text{com}(K,L)$ and

$$I=(K/L)\cup M,\quad J=(L/K)\cup M\,.$$

In the last case, M is a set of $q-d$ indices selected from among the $p-d$ indices common to K and L, so there are $\binom{p-d}{q-d}$ terms in the sum. In connection with the convention on ordering of the sets identified by superscripts on $\underline{B}$, it must be further stipulated that the definitions of I and J are interchanged if necessary so that $I<J$. As indicated previously for pair ordering, $[IJ]$ identifies a pair of index sets $I=(i_1,\ldots,i_p)$ and $J=(j_1,\ldots,j_p)$, with $i_1<\cdots<i_q$ identical with the indices in the set K/L and $i_{q+1}=m_1<\cdots<i_p=m_{p-q}$ being the elements of a particular M, and similarly for J. The order in which the sets are considered to occur does not change if the indices within the sets are reordered. Expressions analogous to those of Eq. (9) could, but will not, be written for the reduction of a $\underline{C}^{[KL]}$.

We see from these results that the image under reduction of a basis element in $\mathcal{E}_p$ is in general a linear combination of basis elements in $\mathcal{E}_q$, and that the preimage in $\mathcal{E}_p$ of a basis element in $\mathcal{E}_q$ would involve a number of the basis elements of $\mathcal{E}_p$ in a rather complicated way. The simple basis set is thus not convenient if we are to consider reduction. We note also that in I we found it necessary to make a change within the $\underline{A}$ basis elements in order to treat the trace condition on a density matrix in a simple way. It will be shown in Sec. III that a particular choice of basis, which includes the $\underline{A}^0$ of paper I as one of its elements, will greatly simplify our description of the reduction process.

III. THE REDUCING BASIS

As the first step in obtaining a more appropriate basis for each $\mathcal{E}_p$, we combine all the $\mathcal{E}_p$ into a

single, larger space

$$\mathcal{E} = \sum_p \oplus \mathcal{E}_p \ . \tag{10}$$

An element in $\mathcal{E}$ can thus have components in any or all of the $\mathcal{E}_p$. Explicit representations can only be constructed as indicated sums, since the addition of matrices having different dimensions is not defined. In practice we will actually have need only for elements entirely within one $\mathcal{E}_p$ but the formal extension is useful. It will present no problems provided we can define a scalar product between elements in different $\mathcal{E}_p$. We do this by requiring that

$$(\underline{U}, \underline{V}) = 0 \ , \quad \underline{U} \in \mathcal{E}_p, \quad \underline{V} \in \mathcal{E}_{p'}, \quad p' \neq p \ . \tag{11}$$

Such a definition will lead to no difficulties: the result is that the $\mathcal{E}_p$ are orthogonal subspaces of $\mathcal{E}$. We will see later that in fact this extension of the scalar product and the orthogonality of the different $\mathcal{E}_p$ are entirely appropriate.

We can now consider the process of reduction to correspond to a *linear operator* acting on any element of $\mathcal{E}$ to produce another element of $\mathcal{E}$ (possibly 0). Rather than considering the general reduction operation $\hat{L}_p^q$, let us consider an operation proportional to the operation of reducing p by 1:

$$\Lambda_- = \binom{p}{p-1} \hat{L}_p^{p-1} = p \, \hat{L}_p^{p-1} \ . \tag{12}$$

It is understood that the p is appropriate to whatever element or component of an element of $\mathcal{E}$ Λ_- is acting on. More precisely, Λ_- could be written as a sum of terms of the form given, each multiplied by a projector onto the corresponding $\mathcal{E}_p$. The space $\mathcal{E}$ is spanned by the totality of the basis elements for the various $\mathcal{E}_p$, and the results of Sec. II give the effect of reduction on any such basis element. The effect of Λ_- on any element in $\mathcal{E}$ is thus determined. Since Λ_- is a well-defined linear operator it will have an *adjoint* $\Lambda_+ \equiv \Lambda_-^\dagger$ such that[5]

$$(\Lambda_+ \underline{U}, \underline{V}) = (\underline{U}, \Lambda_- \underline{V}) \ \text{ all } \underline{U}, \underline{V} \text{ in } \mathcal{E}. \tag{13}$$

The operator Λ_+ will be obtained by postulating an operator through giving its effect on any element of $\mathcal{E}$ and then showing that this operator, as Λ_+, satisfies Eq. (13).

Before doing this, however, we pause to decompose $\mathcal{E}$ into a different set of orthogonal subspaces. For each positive integer $p \leq r$, we choose another integer d such that $0 \leq d \leq \min(p, r-p)$. For each d, then, we can identify a subspace of $\mathcal{E}$. Corresponding to $d = 0$ we define $\mathcal{E}_p(0)$, which is just the subspace of $\mathcal{E}_p$ spanned by the basis elements $\underline{A}^K(p)$ for all $\binom{r}{p}$ choices of K. The dimension of $\mathcal{E}_p(0)$ is this $\binom{r}{p}$. For each $d > 0$ we define two families of subspaces, $\mathcal{E}_p^+(d, \overline{KL})$ and $\mathcal{E}_p^-(d, \overline{KL})$, for integers p between d and $r-d$ and any choice of $\overline{KL}$, where $\overline{KL}$ denotes a pair of *disjoint* sets of d indices each, and the first member of the pair is taken to be the one which comes first in the standard ordering of sets. The space $\mathcal{E}_p^+(d, \overline{KL})$ is the subspace of $\mathcal{E}_p$ spanned by all the basis elements $\underline{B}^{[KL]}(p)$ for which K and L are such that their difference is d and $K/L, L/K$ make up the specified disjoint pair $\overline{KL}$. The space $\mathcal{E}_p^-(d, \overline{KL})$ is similarly defined with $\underline{C}^{[KL]}$ as basis elements. The dimension of $\mathcal{E}_p^+(d, \overline{KL})$ and that of $\mathcal{E}_p^-(d, \overline{KL})$ are each $\binom{r-2d}{p-d}$, the number of ways in which the $p-d$ indices in $\mathrm{com}(K, L)$ can be chosen from among the $r-2d$ indices not in $\overline{KL}$. In the same way that we formally combined the spaces $\mathcal{E}_p$ to form $\mathcal{E}$, we now define

$$\mathcal{E}(0) = \sum_{p=0}^{r} \oplus \mathcal{E}_p(0)$$

$$\mathcal{E}^\pm(d, \overline{KL}) = \sum_{p=d}^{r-2d} \oplus \mathcal{E}_p^\pm(d, \overline{KL}) \ . \tag{14a}$$

These are subspaces of $\mathcal{E}$, and

$$\mathcal{E} = \mathcal{E}(0) \oplus \sum_d \sum_{\overline{KL}} \oplus [\mathcal{E}^+(d, \overline{KL}) \oplus \mathcal{E}^-(d, \overline{KL})] \ . \tag{14b}$$

The sum over d now extends from 1 to $\frac{1}{2}r$, if r is even, or $\frac{1}{2}(r-1)$, if r is odd, and the sum over $\overline{KL}$ ranges over all choices of these disjoint sets.

Any element of $\mathcal{E}_p(0)$ can, by definition, be expressed as a linear combination of $\underline{A}^K(p)$'s. The reduction operator is linear, and we have seen [cf. Eq. (7)] that the result of applying $\hat{L}_p^q$ to any $\underline{A}^K(p)$ is a linear combination of the $\underline{A}^K(q)$'s. It follows that reduction of an element of $\mathcal{E}(0)$ will yield an element of $\mathcal{E}(0)$. A similar result holds for each $\mathcal{E}^\pm(d, \overline{KL})$: Eqs. (9) and the associated discussion show that the result of the application of $\hat{L}_p^q$ to an element of $\mathcal{E}_p^\pm(d, \overline{KL})$ will either be a nonzero element of $\mathcal{E}_q^\pm(d, \overline{KL})$ or the zero element which is common to all subspaces of $\mathcal{E}$.

Within each $\mathcal{E}(d, \overline{KL})$, the indices contained in $\overline{KL}$ are common to all basis elements. The family of spaces $\mathcal{E}_p^+(d, \overline{KL})$ or $\mathcal{E}_p^-(d, \overline{KL})$ for different p but fixed d and $\overline{KL}$ will be isomorphic with a sequence of spaces $\mathcal{F}_p(d)$ independent of $\overline{KL}$. Each $\mathcal{F}_p(d)$ is spanned by an orthonormal set of basis elements $\underline{F}^J(p, d)$ which are analogous to the $\underline{A}$'s, but defined with respect to a set of $p-d$ indices selected from $r-2d$ possible indices. The dimension of $\mathcal{F}_p(d)$ is thus the same as that of $\mathcal{E}_p^+(d, \overline{KL})$ or $\mathcal{E}_p^-(d, \overline{KL})$, and a 1:1 correspondence between basis elements can be established in a way which will be preserved under reduction. One could, for example, delete the indices contained in $\overline{KL}$ from all sets and renumber the remaining indices, in natural order, from 1 through $r-2d$ to go from

$\mathcal{E}_p^{\pm}(d, \overline{KL})$ to $\mathcal{F}_p(d)$. This renumbering is reversible, and the indices within $\overline{KL}$ can be uniquely reinserted in the pair-ordering scheme to establish the correspondence in the other direction. The single (set) index J on $\underline{F}^J$ corresponds to the set com(K, L). We can treat all the spaces $\mathcal{E}^+(d, \overline{KL})$ and $\mathcal{E}^-(d, \overline{KL})$ at the same time by considering a space

$$\mathcal{F}(d) = \sum_p \mathcal{F}_p(d) \tag{15}$$

for general values of d. The allowed range of p is $d \leq p \leq r - d$. We can include $d = 0$, with $\mathcal{F}(0) = \mathcal{E}(0)$.

From the previous expressions and the definitions of Λ_-,

$$\Lambda_- \underline{F}^J(p, d) = \begin{cases} \sum_{k \in J} \underline{F}^{J-k}(p-1, d), & p > d \\ 0, & p = d \end{cases} \tag{16}$$

where $J - k$ denotes the set of indices other than k contained in J. We then define

$$\Lambda_+ \underline{F}^J(p, d) = \begin{cases} \sum_{l \notin J} \underline{F}^{J+l}(p+1, d), & p < r - d \\ 0, & p = r - d \end{cases} \tag{17}$$

where $J + l$ is the set of indices in J and the additional distinct index l.[6] Now let general elements of $\mathcal{F}_p(d)$ be given by

$$\begin{aligned} \underline{U}(p) &= \sum_K u_K \underline{F}^K(p, d), \\ \underline{V}(p) &= \sum_L v_L \underline{F}^L(p, d), \end{aligned} \tag{18}$$

where the coefficients u_K and v_L are real numbers. The two cases covered in each of Eqs. (16) and (17) can be treated at once by the use of Kronecker deltas. We then find

$$\begin{aligned} &(\underline{U}(p), \Lambda_- \underline{V}(p')) \\ &= \sum_{K,L} (1 - \delta_{p',d}) \sum_{l \in L} (\underline{F}^K(p, d), \underline{F}^{L-l}(p' - 1, d)) \\ &= (1 - \delta_{p',d}) \delta_{p,p'-1} \sum_L \sum_{l \in L} u_{L-l} v_L \\ &= \delta_{p,p'-1} \sum_L \sum_{l \in L} u_{L-l} v_L . \end{aligned} \tag{19}$$

We can drop the $\delta_{p',d}$ to obtain the last equality because $p \geq d$ so if $p' = d$, $\delta_{p,p'-1} = 0$ for all allowed p. Similarly,

$$\begin{aligned} (\Lambda_+ \underline{U}(p), \underline{V}(p')) &= \sum_{K,L} u_K v_L (1 - \delta_{p,r-d}) \\ &\quad \times \sum_{j \in K} (\underline{F}^{K+j}(p+1, d), \underline{F}^L(p', d)) \\ &= (1 - \delta_{p,r-d}) \delta_{p+1,p'} \sum_K \sum_{j \in K} u_K v_{K+j} \\ &= \delta_{p+1,p'} \sum_K \sum_{j \in K} u_K v_{K+j} \\ &= \delta_{p,p'-1} \sum_L \sum_{l \in L} u_{L-l} v_L \\ &= (\underline{U}(p), \Lambda_- \underline{V}(p')) \end{aligned} \tag{20}$$

Any element of $\mathcal{F}(d)$ can be resolved into components in the various $\mathcal{F}_p(d)$, and the results above show that Eq. (13) will be satisfied in the general case, so $\Lambda_+ = \Lambda_-^\dagger$.

We next consider the commutator of Λ_+ and Λ_-. We begin by examining the action of the products $\Lambda_+\Lambda_-$ and $\Lambda_-\Lambda_+$ on a basis element of $\mathcal{F}_p(d)$:

$$\begin{aligned} \Lambda_+\Lambda_- \underline{F}^K(p, d) &= \Lambda_+ \left((1 - \delta_{p,d}) \sum_{k \in K} \underline{F}^{K-k}(p-1, d) \right) \\ &= (1 - \delta_{p,d})(1 - \delta_{p-1,r-d}) \sum_{k \in K} \sum_{l \notin K-k} \underline{F}^{K-k+l}(p, d) \\ &= (1 - \delta_{p,d}) \left[(p - d) \underline{F}^K(p, d) + \sum_{k \in K} \sum_{l \notin K} F^{K-k+l}(p, d) \right] . \end{aligned} \tag{21}$$

The $\delta_{p-1,r-d}$ would be nonzero only if $p = r - d + 1$, which is not allowed, so it can be dropped. The first term in the large square brackets comes from the $p - d$ terms in the previous double sum for which $k = l$, and the remaining sum in brackets consists of the rest of the terms. In a similar way we find

$$\begin{aligned} \Lambda_-\Lambda_+ \underline{F}^K(p, d) &= \Lambda_- \left((1 - \delta_{p,r-d}) \sum_{l \notin K} \underline{F}^{K+l}(p+1, d) \right) \\ &= (1 - \delta_{p,r-d})(1 - \delta_{p+1,d}) \sum_{l \notin K} \sum_{k \in K+l} F^{K+l-k}(p, d) \\ &= (1 - \delta_{p,r-d}) \left([r - 2d - (p - d)] \underline{F}^K(p, d) + \sum_{k \in K} \sum_{l \notin K} \underline{F}^{K-k+l}(p, d) \right) . \end{aligned} \tag{22}$$

The action of the commutator $[\Lambda_+, \Lambda_-]$ on $\underline{F}^K(p,d)$ is thus

$$(\Lambda_+\Lambda_- - \Lambda_-\Lambda_+)\underline{F}^K(p,d) = \{(p-d) - [r-2d-(p-d)]\}\underline{F}^K(p,d) + \{\delta_{p,d}(p-d) - \delta_{p,r-d}[r-2d-(p-d)]\} \times \underline{F}^K(p,d) - (\delta_{p,d} - \delta_{p,r-d}) \sum_{k\in K} \sum_{l\notin K} \underline{F}^{K-k+l}(p,d) = (2p-r)\underline{F}^K(p,d). \quad (23)$$

The expression in curly brackets in the second term is zero because the coefficient of each Kronecker delta is zero for the value of p which makes the delta nonzero, and the term involving summations is in fact zero because if $p=d$ there can be no $k\in K$, while if $p=r-d$ there can be no $l\in K$. We have seen that $[\Lambda_+, \Lambda_-]$ acting on any basis element $\underline{F}^K(p,d)$ produces a multiple of that basis element, and the coefficient is independent of K, so the same result will hold for any general element of $\mathfrak{F}_p(d)$. More generally, for any $\underline{U}$ in $\mathfrak{F}(d)$,

$$\underline{U} = \sum_{p,K} u_{p,K}\underline{F}^K(p,d), \quad (24)$$

we have

$$[\Lambda_+, \Lambda_-]\underline{U} = \sum_{p,K} u_{p,K}(-\tfrac{1}{2}r+p)\underline{F}^K(p,d). \quad (25)$$

With this incentive we define

$$\Lambda_1 = \tfrac{1}{2}(\Lambda_+ + \Lambda_-), \qquad \Lambda_2 = -\tfrac{1}{2}i(\Lambda_+ - \Lambda_-), \quad (26)$$

and

$$\Lambda_3 = \tfrac{1}{2}[\Lambda_+, \Lambda_-]. \quad (27)$$

It follows immediately from these definitions and the fact that $\Lambda_+ = \Lambda_-^\dagger$ that these new operators are self-adjoint and that

$$[\Lambda_1, \Lambda_2] = i\Lambda_3. \quad (28)$$

From the effect of Λ_+ and Λ_-, given in Eqs. (16) and (17), and that of Λ_3, corresponding to Eq. (23), we find that

$$[\Lambda_3, \Lambda_+] = \Lambda_+, \qquad [\Lambda_3, \Lambda_-] = -\Lambda_-, \quad (29)$$

so that, since $\Lambda_\pm = \Lambda_1 \pm i\Lambda_2$,

$$[\Lambda_3, \Lambda_1] = i\Lambda_2, \qquad [\Lambda_3, \Lambda_2] = -i\Lambda_1. \quad (30)$$

These relationships follow most directly for the action of the operators on basis elements $\underline{F}^K(p,d)$, but since they hold for all basis elements (choices of p and K), they hold for the operators acting on general elements of the space $\mathfrak{F}(d)$. The operators Λ_1, Λ_2, and Λ_3 may thus be thought of as the components of an *angular momentum* Λ.[7] Any one component, in particular Λ_3, will commute with

$$\Lambda^2 = \Lambda_1^2 + \Lambda_2^2 + \Lambda_3^2. \quad (31)$$

The basis elements $\underline{F}^K(p,d)$ are eigenvectors of Λ_3 but not of Λ^2. We can define new basis elements for $\mathfrak{F}_p(d)$, by an orthogonal transformation of the $\underline{F}^K(p,d)$, which will be eigenvectors of Λ^2.

The easiest way of obtaining such basis elements for $\mathfrak{F}_p(d)$ will be seen if we note an analogy between the $\underline{F}^K(p,d)$ and simple spin-product functions. A basis element $\underline{F}^K(p,d)$ is determined by specifying a set of $p-d$ indices from among $r-2d$ possible indices. There is clearly a one-to-one correspondence between such a selection of indices and a product of $r-2d$ spin-$\frac{1}{2}$ functions α or β, with the function in position k being α if $k\in K$ and β if $k\notin K$. This function is an eigenfunction of S_z with eigenvalue $\frac{1}{2}[(p-d)-(r-2d-p+d)] = -\frac{1}{2}r+p$. The number of such product functions with $p-d$ α's is $\binom{r-2d}{p-d}$ and the total number of spin-product functions for $r-2d$ spin-$\frac{1}{2}$ particles is 2^{r-2d}. These are the same as the dimensions of $\mathfrak{F}_p(d)$ and $\mathfrak{F}(d)$, respectively. This analogy becomes an isomorphism when we note that the effect of $S_\pm$ on one of the spin-product functions is the same as the effect of $\Lambda_\pm$ on the corresponding $\underline{F}^K(p,d)$. We can therefore obtain eigenvectors of Λ^2 by applying to the $\underline{F}^K(p,d)$ the same orthogonal transformation which converts the spin-product functions into S^2 eigenfunctions. This transformation has been extensively studied and can be accomplished in a variety of well-known ways.

A new, complete, orthonormal basis for $\mathcal{S}$ is now definable. The space is first decomposed into a series of orthogonal subspaces $\mathcal{S}(d, \overline{KL})$. In each of these subspaces an identification is made between basis elements of the $\underline{A}$ type (if $d=0$) or $\underline{B}$ and $\underline{C}$ types (for $d>0$) and spin-product functions for $r-2d$ spin-$\frac{1}{2}$ particles. The basis elements are then subjected to the same orthogonal transformation which, when applied to the spin-product functions, would result in spin eigenfunctions. The result will be basis elements for $\mathcal{S}(0)$, $\mathcal{S}^+(d, \overline{KL})$, or $\mathcal{S}^-(d, \overline{KL})$ which are eigenvectors of Λ^2 and Λ_3. Let the eigenvalues associated with a particular basis element be denoted by

$\lambda(\lambda+1)$ and μ, respectively. We already know that μ is related to p by $\mu=-\frac{1}{2}r+p$. Unless otherwise specified, we will now assume that $\mu\leq 0$, $p\leq\frac{1}{2}r$. This will be the case normally, since the largest value of p we will be interested in is n, the number of electrons in a system of interest, and r, the number of basis spin orbitals is nearly always at least $2n$. We can also relate λ to another quantity of more direct interest. As Λ_- is applied to the basis element, the value of μ is decreased until the limiting value of $-\lambda$ is attained. Further application of Λ_- will then give zero. We can thus relate λ to the minimum value of p for which this particular basis element can exist. Let us use π to denote this minimum value of p. Then

$$-\tfrac{1}{2}r+\pi=-\lambda\,,\quad \lambda=\tfrac{1}{2}r-\pi\,. \tag{32}$$

Since the maximum possible value of λ is $\frac{1}{2}(r-2d)$, the minimum value for π is d. This is consistent with the intended meaning of π, since the lowest value of p for which a basis element can possibly exist is equal to the d value for that basis element. The new basis elements will be denoted by $\underline{X}$, $\underline{Y}$, and $\underline{Z}$ with the following symbolic relation to the $\underline{A},\underline{B},\underline{C}$ basis:

$$\begin{aligned}&\{\underline{A}^K(p)\}\rightarrow\{\underline{X}_{\pi g}(p)\}\,,\\ &\{\underline{B}^{KL}(p)\}\rightarrow\{\underline{Y}_{\pi g}(p,d,\overline{KL})\}\,,\\ &\{\underline{C}^{KL}(p)\}\rightarrow\{\underline{Z}_{\pi g}(p,d,\overline{KL})\}\,.\end{aligned} \tag{33}$$

The additional index g is a "spin degeneracy" index with the range[8]

$$1\leq g\leq f(r-2d,\lambda)=\binom{r-2d}{\pi-d}-\binom{r-2d}{\pi-d-1}(1-\delta_{\pi,d})\,. \tag{34}$$

It serves to distinguish basis elements with the same values of λ or π and μ or p, as well as common $d,\overline{KL}$. When $d=0$, the $\underline{A}$ basis is involved and can be converted to an $\underline{X}$ basis. Note that $\underline{X}$ and $\overline{A}^0$ of I are in fact eigenfunctions of Λ^2 with $\lambda=\frac{1}{2}r$ and $\overline{A}^0$ thus corresponds to $\underline{X}_{01}(p)$ in the present notation. The spaces spanned by the $\underline{B}$'s or the $\underline{C}$'s are orthogonal subspaces of each $\mathcal{S}_p$ which can be treated separately so that the identities of $\underline{Y}$ and $\underline{Z}$ are preserved. In particular, if one is dealing with real matrices, $\underline{C}$'s or $\underline{Z}$'s can be omitted.

The new basis has particularly simple behavior under reduction, and for that reason it will be called the *reducing basis*.

Let $\underline{G}_{\pi g}(p,d,\overline{KL})$ be one of $\underline{X}$, $\underline{Y}$, or $\underline{Z}$, with $d=0$ and $\overline{KL}$ understood to be omitted for an $\underline{X}$. From the way in which these elements were constructed and the relationship between the reduction operation and Λ_-,

$$\hat{L}_p^q\underline{G}_{\pi g}(p,d,\overline{KL})=\begin{cases}\zeta(p,q,\pi)\underline{G}_{\pi g}(q,d,\overline{KL}) & \text{if } q\geq\pi\\ 0 & \text{if } q<\pi\,,\end{cases} \tag{35}$$

where

$$\zeta(p,q,\pi)=\frac{[(p-\pi)(p-1-\pi)\cdots(q+1-\pi)]^{1/2}[(r-\pi-p+1)(r-\pi-p+2)\cdots(r-\pi-q)]^{1/2}}{p(p-1)\cdots(q+1)}$$
$$=\frac{q!}{p!}\left(\frac{(p-\pi)!}{(q-\pi)!}\,\frac{(r-\pi-q)!}{(r-\pi-p)!}\right)^{1/2}. \tag{36}$$

It is well known that the spin raising and lowering operators leave unchanged both the total-spin quantum number S and the spin-degeneracy label. Since an isomorphism has been established between that case and the present case, Λ_- or $\hat{L}_p^q$ will not change π and g. The coefficient arises from the conversion of $(\Lambda_-)^{p-q}$ to $\hat{L}_p^q$ and the usual angular momentum factor corresponding to $[S(S+1)-M(M-1)]^{1/2}$. We have thus established a one-to-one correspondence between basis elements in $\mathcal{S}_p$ and $\mathcal{S}_q$, for any p and $q<p$, except that some of the basis elements of $\mathcal{S}_p$ will map into the zero-element of $\mathcal{S}_q$. Given any element of $\mathcal{S}_p$ expressed in the reducing basis, we can immediately determine what its image in $\mathcal{S}_q$ will be. Further, given any element in $\mathcal{S}_q$, its preimage in $\mathcal{S}_p$ expressed in the reducing basis will have some components completely fixed and others (those with $\pi>q$) completely arbitrary.

The reduction of $\mathcal{S}_p$ onto $\mathcal{S}_q$ can also be thought of as a projection (with scaling) of $\mathcal{S}_p$ onto the subspace of $\mathcal{S}_p$ spanned by those basis elements with $\pi\leq q$. Since the coefficient in Eq. (35) depends on π, the scaling is not the same for all directions. Each $\mathcal{S}_p$ can be divided into orthogonal subspaces $\mathcal{S}_{p\pi}$ spanned by those basis elements of $\mathcal{S}_p$ having the specified value of π. Correspondingly, any element $\underline{V}^{(p)}$ in $\mathcal{S}_p$ can be expressed as

$$\underline{V}^{(p)}=\sum_{\pi=0}^{p}\underline{V}^{(p,\pi)}, \tag{37}$$

where

$$\hat{L}_p^q\underline{V}^{(p,\pi)}=\begin{cases}\underline{V}^{(q,\pi)} & \text{if } q\geq\pi\\ 0 & \text{if } q<\pi\,.\end{cases} \tag{38}$$

Coleman has introduced a decomposition, based on group theory, of a two-electron reduced density matrix into zero-, one-, and two-electron components.[9] The present decomposition generalizes his result and reduces to it when $p=2$ and $\underline{V}$ is a reduced density matrix. The general group-theoretic decomposition has also been discussed by Coleman,[10] and it seems likely that the group theoretic arguments could be extended to show that each $\mathcal{E}_{p\pi}$ is invariant with respect to a unitary transformation of the spin orbital basis.

Consider now some fixed positive integer n less than r. Any unit-trace element in $\mathcal{E}_n$ can be expressed as $\underline{X}^{(n)}+\underline{V}^{(n)}$, where

$$\underline{X}^{(n)}=\binom{R}{n}^{-1/2}X_{0,1}(n) \tag{39}$$

and $\underline{V}^{(n)}$ is an element of $\mathfrak{N}_n$ (this space is defined in I). For each $p<n$, $\underline{V}^{(n)}$ can be further decomposed as

$$\underline{V}^{(n)}=\underline{V}(n,p)+\underline{V}_{\perp}(n,p)\,, \tag{40}$$

where $\underline{V}(n,p)$ is a linear combination of those reducing basis elements of $\mathfrak{N}_n$ having $\pi\leq p$ and $\underline{V}_{\perp}(n,p)$ is a linear combination of those reducing basis elements of $\mathfrak{N}_n$ having $\pi>p$. From the definitions we have

$$\hat{L}_n^p\underline{X}^{(n)}=\underline{X}^{(p)} \tag{41}$$

and further

$$\begin{aligned}&\hat{L}_n^p\underline{V}(n,p)=\underline{V}(p)\,,\\ &\hat{L}_n^p\underline{V}_{\perp}(n,p)=0\,,\end{aligned} \tag{42}$$

where $\underline{X}^{(p)}+\underline{V}^{(p)}$ is the image under reduction of $\underline{X}^{(n)}+\underline{V}^{(n)}$. We note that the distance of this original element from $\underline{X}^{(n)}$ is just the norm of $\underline{V}^{(n)}$ and further that

$$\|\underline{V}^{(n)}\|=\|\underline{V}(n,p)\|+\|\underline{V}_{\perp}(n,p)\| \tag{43}$$

since the components are in orthogonal subspaces.

For any $p<n\leq\frac{1}{2}r$ we can define a new operator $\hat{\Gamma}_p^n$ as[11]

$$\begin{aligned}\hat{\Gamma}_p^n\underline{V}(p)=&\text{ that element of the subspace }\sum_{\pi=1}^{p}\oplus\,\mathcal{E}_n\\ &\text{such that, when }\hat{L}_n^p\text{ is applied to it,}\\ &\underline{V}(p)\text{ results.}\end{aligned} \tag{44}$$

Equivalently, if $\underline{V}(p)$ is expressed in terms of the reducing basis elements, $\hat{\Gamma}_p^n\underline{V}(p)$ will have the same expansion coefficients in terms of those basis elements of $\mathfrak{N}_n$ having $\pi\leq p$. It follows that

$$\hat{L}_n^p\hat{\Gamma}_p^n\underline{V}(p)=\underline{V}(p) \tag{45}$$

for any $\underline{V}(p)$ in $\mathfrak{N}_p$. It is not true, however, that $\hat{\Gamma}_p^n\hat{L}_n^p\underline{V}(n)=\underline{V}(n)$ for general elements of $\mathfrak{N}_n$, since all information about $\underline{V}_{\perp}(n,p)$ is lost. While $\hat{\Gamma}_p^n$ functions as a right inverse of $\hat{L}_n^p$, in fact $\hat{L}_n^p$ is a projector and as such cannot have an inverse.

IV. N REPRESENTABILITY

The treatment of Hermitian, unit-trace matrices as elements in a vector space suggests some approaches to the N-representability problem. A necessary and a sufficient condition are readily obtained. A restatement and possibly a formal solution to the problem also result. It will be useful to begin this section with a brief discussion of what really constitutes a "solution" to the N-representability problem.

The N-representability problem can be stated as follows: give necessary and sufficient conditions (NASC) on a (Hermitian, unit-trace) operator $D^{(p)}$ such that it is obtainable by reduction of an n-particle density matrix [as in Eq. (2)]. More stringently, the n-particle density matrix may be required to be a pure state, in which case $D^{(p)}$ is obtainable from a wave function (antisymmetric).[12] This is the pure-state N-representability problem. Various "solutions" to these problems have been proposed, often involving a construction procedure which, if N representability is possible, will produce a wave function or n matrix.[13,14] Others involve still more complex processes.[2,15]

The following terminology is proposed to categorize "solutions" to the N-representability problem:

(i) A *restatement* of the problem. When obtaining NASC for N representability is shown to be equivalent to the solution of some other problem, but where a (finite) algorithm for the solution of that problem is not available.

(ii) A *formal solution* of the problem. When a finite algorithm is presented whereby the N representability of a given p matrix can be tested or the class of N-representable p matrices can be parametrized or defined by constraints, but where the effort involved in testing or in doing a variational calculation with the parametrized or constrained p matrix is comparable to or greater than the effort required to obtain the lowest eigenvalue and corresponding eigenvector of a full configuration-interaction (CI), n-electron Hamiltonian matrix.

(iii) A *practical solution* of the problem. When an algorithm to test N representability of a parametrized or constrained p matrix is presented so that the reduced density matrix can be obtained directly with distinctly less effort than would be involved in the determination of a wave function or n-matrix for the same system and state.

It is also possible to deal with *approximate* N representability. Typically, constraints which are known to be necessary but not sufficient for N

representability are imposed and one hopes for the best. Although confidence in the utility of this approach may be developed on the basis of experience,[16-20] this will take time and is not entirely satisfactory. It would be preferable to introduce some measure of how nearly N representable a p matrix is, and to develop bounds, in terms of this measure, on the errors which may result as a consequence of non-N-representability.[21] The geometric formulation provides a way to do the former and may suggest an approach to the latter.

For any density matrix $\underline{D}^{(p)}$ in $\mathcal{S}_p$ we define a subset of $\mathfrak{N}_n$, $\mathcal{G}_n(\underline{D}^{(p)})$ $(n>p)$ which we will call the preimage of $\underline{D}^{(p)} - \underline{X}^{(p)}$.[22] It is defined as the set of all elements $\underline{V}^{(n)}$ in $\mathfrak{N}_n$ such that

$$\hat{L}_n^p \underline{V}^{(n)} = \underline{D}^{(p)} - \underline{X}^{(p)} \tag{46}$$

or, equivalently,

$$\hat{L}_n^p (\underline{V}^{(n)} + \underline{X}^{(n)}) = \underline{D}^{(p)} . \tag{47}$$

Using the notation defined at the end of the previous section, we decompose $\underline{V}^{(n)}$ into

$$\underline{V}^{(n)} = \underline{V}(n,p) + \underline{V}_\perp(n,p) \tag{48}$$

and note that

$$\underline{V}(n,p) = \hat{\Gamma}_p^n D^{(p)} \tag{49}$$

is completely determined by $\underline{D}^{(p)}$ while $\underline{V}_\perp(n,p)$ is completely arbitrary. The component $\underline{V}(n,p)$ will be referred to as the minimal-norm preimage element of $\underline{D}^{(p)}$.

With this background, we now consider the following restatement of the N-representability problem: *A p-electron density matrix $\underline{D}^{(p)}$ is N-representable if and only if its preimage in $\mathfrak{N}_n$ intersects $\mathcal{P}^{(n)} - \underline{X}^{(n)}$, i.e., $\mathcal{G}_n(D^{(p)}) + \underline{X}^{(n)}$ includes a positive element.* In the present formulation, Hermiticity and antisymmetry are assured. Use of the reducing basis greatly simplifies the specification of the preimage and takes care of the trace condition. All the difficulty is now associated with the determination of positivity. If a sufficiently convenient characterization of $\mathcal{P}^{(n)}$ could be given, a solution to the N-representability problem would result.

In order to transform this restatement of the problem into a solution, a means of testing must be provided. Any single element in the preimage can in principle be tested for the positivity of its sum with $\underline{X}^{(n)}$. Thus, for example, the positivity of $\underline{X}^{(n)}$ plus the preimage element of minimal norm can be tested. Such positivity provides a sufficient condition for N representability. If it could be shown that positivity of $X^{(n)}$ plus any preimage element implies positivity of $X^{(n)}$ plus the minimal norm preimage element, this condition would also be necessary. However, simple examples indicate that such is not the case.

This result is probably related to Coleman's formal solution of the N-representability problem,[9,23] but a complete connection has not yet been established.

Another sufficient condition and a necessary condition can be stated in terms of the norm of the minimal-norm preimage element $f(D^{(p)}) = \|V(n,p)\|$, and the radii of the inner and outer hyperspheres in $\mathfrak{N}_n$: *If $f(D^{(p)}) < d_{\min}^{(n)} = \{[\binom{r}{n} - 1]\binom{r}{n}\}^{-1/2}$ then $\underline{D}^{(p)}$ is N representable. If $f(D^{(p)}) > d_{\max}^{(n)} = \{[\binom{r}{n} - 1]/\binom{r}{n}\}^{1/2}$, $\underline{D}^{(p)}$ is not N representable.* These conditions could also be stated in terms of the components of $\underline{D}^{(p)}$ directly, by taking into account the scaling associated with reduction.

In order to apply these results (from I) we need to be able to relate the norm of an element to the norm of its image under reduction. Suppose that an element $\underline{V}^{(n)}$ of $\mathfrak{N}_n$ is given. We consider now the determination of the norm of $\hat{L}_n^p \underline{V}^{(n)}$. This is easily done if $\underline{V}^{(n)}$ is given in terms of the reducing basis. It can also be done if $\underline{V}^{(n)}$ is given in terms of the $\underline{A},\underline{B},\underline{C}$ basis without the need for a transformation to the reducing basis.

If $\underline{V}^{(n)}$ is given in terms of the reducing basis we can immediately make the decomposition as in Eq. (37),

$$\underline{V}^{(n)} = \sum_{\pi=1}^{n} \underline{V}(n,\pi)$$

and note that

$$\hat{L}_n^p \underline{V}^{(n)} = \sum_{\pi=1}^{p} \hat{L}_n^p \underline{V}(n,\pi) . \tag{50}$$

From Eq. (35), then

$$\| \hat{L}_n^p \underline{V}(n,\pi) \| = \zeta(n,p,\pi) \| V(n,\pi) \| , \tag{51}$$

and since components with different values of π are orthogonal,

$$\| \hat{L}_n^p \underline{V}^{(n)} \| = \sum_{\pi=1}^{p} \zeta(n,p,\pi) \| \underline{V}(n,\pi) \| . \tag{52}$$

When $\underline{V}^{(n)}$ is given in terms of the simple $\underline{A},\underline{B},\underline{C}$ basis and we wish to avoid the necessity of transforming to the reducing basis, each basis element (and thus $\underline{V}^{(n)}$) can be decomposed into components of definite π by the use of a projection operator analogous to the Löwdin spin-projection operator.[24] Norms of these components will be given in terms of Sanibel coefficients.[25-28] Let

$$\underline{V}^{(n)} = \sum_{t=-n}^{n} \sum_{\overline{KL}} \sum_{J} u(t,\overline{KL},J) \underline{U}(n,t,\overline{KL},J)$$

$$\underline{U}(n,t,\overline{KL},J) = \begin{cases} \underline{A}^J & \text{if } t=0 \quad (\overline{KL} \text{ omitted}) \\ \underline{B}^{KL} & \text{if } t>0 \\ \underline{C}^{KL} & \text{if } t<0 \end{cases} \Bigg\} \; KL = \overline{KL} \cup J , \tag{53}$$

where $\overline{KL}$ represents a set of $2|t|$ indices, the first $|t|$ in K/L and the second $|t|$ in L/K, J represents a set of $n-|t|$ indices chosen from the $r-2|t|$ indices not in $\overline{KL}$, and u is an expansion coefficient. Since $\hat{L}_n^p \underline{U}(n,t,KL,J)$ is orthogonal to $\hat{L}_n^p \underline{U}(n,t',\overline{KL}',J')$ unless $t'=t$ and $\overline{KL}'=\overline{KL}$,

$$\| \hat{L}_n^p \underline{V}^{(n)} \|^2 = \sum_t \sum_{\overline{KL}} x(t,\overline{KL}), \tag{54}$$

where

$$x(t,\overline{KL}) = \sum_{J,J'} u(t,\overline{KL},J)u(t,\overline{KL},J')\Delta(J,J'), \tag{55}$$

$$\Delta(J,J') = (\hat{L}_n^p \underline{U}(n,t,\overline{KL},J),\ \hat{L}_n^p \underline{U}(n,t,\overline{KL},J')).$$

Now introduce projection operators Θ_π, projecting elements of $\mathcal{S}$ with the given values of $d=|t|$ onto the subspace of Λ^2 eigenfunctions with eigenvalue $\lambda=\frac{1}{2}r-\pi$. Since the sum of Θ_π over all allowed values of π provides a resolution of the identity and projected functions with different values of π will be orthogonal,

$$\begin{aligned}\Delta(J,J') &= \Big(\hat{L}_n^p \sum_\pi \Theta_\pi \underline{U}(n,t,\overline{KL},J),\\ &\qquad \hat{L}_n^p \sum_{\pi'} \Theta_{\pi'} \underline{U}(n,t,\overline{KL},J')\Big)\\ &= \sum_{\pi=\max(d,1)}^{p} (\hat{L}_n^p \Theta_\pi \hat{P}(J)\underline{U}_0,\ \hat{L}_n^p \Theta_\pi \hat{P}(J')\underline{U}_0).\end{aligned} \tag{56}$$

We have introduced here the particular element $\underline{U}_0$ (an $\underline{A}$ if $t=0$, $\underline{B}$ if $t>0$, or $\underline{C}$ if $t<0$) in which the indices included in the J group are the first $n-d$ from the set of $r-2d$ available for the given $\overline{KL}$, and $\hat{P}(J)$ is an operator such that

$$\underline{U}(n,t,\overline{KL},J) = \hat{P}(J)\underline{U}_0. \tag{57}$$

In the analogous spin-function case, $\underline{U}_0$ becomes $[\alpha^{n-d}|\beta^{r-d-n}]$ and $\hat{P}(J)$ is a permutation operator.[29] The sum over π values can be truncated because components with $\pi>p$ will not survive reduction and no element of $\mathfrak{N}$ can have a component with $\pi=0$.

We use the relationship between $\hat{L}_n^p$ and $(\Lambda_-)^{n-p}$ implied by Eq. (12) and the properties of angular momentum operators to obtain, as in Eq. (35),

$$\begin{aligned}&(\hat{L}_n^p \Theta_\pi \hat{P}(J)\underline{U}_0,\ \hat{L}_n^p \Theta_\pi \hat{P}(J')\underline{U}_0)\\ &\quad = [\zeta(n,p,\pi)]^2 (\Theta_\pi \hat{P}(J)\underline{U}_0, \Theta_\pi \hat{P}(J')\underline{U}_0)\\ &\quad = [\zeta(n,p,\pi)]^2 ([\hat{P}(J')]^{-1}\hat{P}(J)\underline{U}_0, \Theta_\pi \underline{U}_0).\end{aligned} \tag{58}$$

The second equality is a consequence of the fact that the projection operator Θ_π is Hermitian and idempotent, and commutes with $\hat{P}(J)$, while $\hat{P}(J')$ is unitary. These properties are well known in the analogous spin projection case, and could readily be proved here. Now, define $[\underline{U}]_k$ as the sum of all elements $\underline{U}(n,t,\overline{KL},J)$ for which the index set J contains k of the last $r-d-n$ of the available indices. (It is analogous to the sum of spin-product functions $[\alpha^{n-d-k}\beta^k|\alpha^k\beta^{r-d-n-k}]$.)

From the theory of spin projections we know that[24,29]

$$\Theta_\pi \underline{U}_0 = \sum_{k=0}^{n-d} C_k(\tfrac{1}{2}r-\pi, \tfrac{1}{2}r-n, \tfrac{1}{2}r-d)[\underline{U}]_k, \tag{59}$$

where C_k is the Sanibel coefficient. (Since the usual spin-projection result assumes $M_S \geq 0$, the sign of μ has been changed and the roles of α and β interchanged to obtain this result.) The single scalar product in Eq. (57) will be replaced by a sum over k, and in each term the scalar product will be zero unless $[\hat{P}(J')]^{-1}\hat{P}(J)\underline{U}_0$ is included in $[\underline{U}]_k$. This will happen for one and only one value of k, so

$$\begin{aligned}&(\hat{L}_n^p \Theta_\pi \hat{P}(J)\underline{U}_0,\ \hat{L}_n^p \Theta_\pi \hat{P}(J')\underline{U}_0)\\ &\quad = [\zeta(n,p,\pi)]^2 C_j(\tfrac{1}{2}r-\pi, \tfrac{1}{2}r-n, \tfrac{1}{2}r-d),\end{aligned} \tag{60}$$

where j is the number of indices from the last $r-d-n$ available included in $[\hat{P}(J')]^{-1}\hat{P}(J)\underline{U}_0$. It is equal to the number of indices differing in the sets J and J'. Finally, then

$$\begin{aligned}\| \hat{L}_n^p \underline{V}^{(n)} \|^2 &= \sum_{t=-n}^{n} \sum_{\overline{KL}} \sum_{J,J'} u(t,\overline{KL},J)u(t,\overline{KL},J')\\ &\quad \times \sum_{\max(d,1)}^{k} [\zeta(n,p,\pi)]^2\\ &\quad \times C_j(\tfrac{1}{2}r-\pi, \tfrac{1}{2}r-n, \tfrac{1}{2}r-d),\end{aligned} \tag{61}$$

where $j=j(J,J')$ and $d=|t|$.

As a special case, we consider $\| \hat{L}_n^1 \underline{A}^K(n) - \underline{X}^{(n)} \|$, i.e., $u=\delta_{d,0}\delta_{JK}$. There is then no sum over $\overline{KL}$, the sum over π includes only one term, $\pi=1$, and $j=0$. Thus,

$$\begin{aligned}&\| \hat{L}_n^1(\underline{A}^K(n) - \underline{X}^{(n)}) \|^2\\ &\quad = [\zeta(n,1,1)]^2 C_0(\tfrac{1}{2}r-1, \tfrac{1}{2}r-n, \tfrac{1}{2}r)\\ &\quad = \frac{(1!)^2}{(n!)^2}\left(\frac{(n-1)!\,(r-2)!}{0!\,(r-n-1)!}\right)(r-1)\,\frac{(r-n)!\,n!}{1!\,r!}\\ &\quad = \frac{r-n}{rn}.\end{aligned} \tag{62}$$

This is of course just the result obtained in I for a Slater 1 matrix.

The geometric formulation leads to a natural measure of approximate N representability. The symbol ξ_n will be used for this quantity,[30] which is defined for any p-electron density matrix $\underline{D}^{(p)}$ by

$$\xi_N(\underline{D}^{(p)}) = d(g_n(\underline{D}^{(p)}), \mathcal{P}^{(n)} - \underline{X}^{(n)}), \tag{63}$$

i.e., the distance between the preimage set of $\underline{D}^{(p)}$

$-\underline{X}^{(p)}$ and the set of $\underline{D}^{(n)}-\underline{X}^{(n)}$. This is defined in the usual way as

$$d(\mathcal{G}_n(\underline{D}^{(p)}),\mathcal{P}^{(n)}-\underline{X}^{(n)})=\min d_{\underline{V}\underline{W}},$$
$$\text{all } \underline{V} \text{ in } \mathfrak{N}_n \text{ such that } \hat{L}_n^p(\underline{V}+\underline{X}^{(p)})=\underline{D}^{(p)}, \quad (64)$$
$$\text{all } \underline{W} \text{ in } \mathfrak{N}_n \text{ such that } \underline{W}+\underline{X}^{(n)}\geq 0,$$

where $d_{\underline{V}\underline{W}}$ is the distance defined in I. The possibility of relating ξ_n to error bounds on properties calculated using $\underline{D}^{(p)}$ will be considered in a later paper.

Another measure could be introduced in terms of the distance of $\underline{D}^{(p)}$ from the set $\mathcal{P}_p^{(n)}$ of N-representable p matrices. It is less satisfactory than ξ_n, however, because $\mathcal{P}_p^{(n)}$ is not fully characterized while $\mathcal{G}_n(\underline{D}^{(p)})$ and $\mathcal{P}^{(n)}$ are, at least in principle.

In this paper, we have seen that a geometric formulation together with an analogy to the theory of spin eigenfunctions provides a new way of looking at the N-representability problem. A restatement of the problem results, and some conditions for N representability and a measure of approximate N-representability have been obtained in terms of distances.

The transformation to the reducing basis in each $\mathcal{E}_n$ is straightforward in principle. Analogous problems have been extensively studied. In practice a calculation of very substantial magnitude would be required, but it need be done only once for a given r and n, independent of the particular system being considered. If a sufficiently convenient characterization of $\mathcal{P}^{(n)}$ could be found, a practical solution of the N-representability problem would result.

The results obtained here also suggest a means of exploring the relationship between the matrix of the reduced Hamiltonian and the full CI Hamiltonian matrix. This relationship will be more fully explored in a later paper, where the important aspects associated with the true spin symmetry of the system are considered.

ACKNOWLEDGMENT

This work was supported by the National Science Foundation under Grant No. CHE 74-17494-A01.

[1]J. E. Harriman, preceding paper, Phys. Rev. A 17, 1249 (1978).
[2]H. Kummer, J. Math. Phys. 8, 2063 (1967).
[3]R. M. Erdahl and C. Garrod, in *Reduced Density Operators with Applications to Physical Systems—II, Queen's Papers in Pure and Applied Mathematics No. 40*, edited by R. M. Erdahl (Queen's University, Kingston, Ontario, 1974), p. 22. Cf. also other papers of Erdahl in the same volume.
[4]The notation is based on that of Kummer (Ref. 2), but the operator defined here is distinguished from that of Kummer by a circumflex. The properties of the operators used here are those which follow from the definition given, and complete equivalence with Kummer's operator is not necessarily assumed.
[5]This corresponds to Kummer's Γ_n^{n+1}.
[6]If the indices in J are thought of as being selected from an original set of r indices with the indices in K/L and L/K deleted, then it is necessary to specify $l\notin K/L$, $l\notin L/K$; if the $r-2d$ indices available for elements of $\mathcal{F}(d)$ are thought of as having an independent existence, such a restriction is not necessary or meaningful.
[7]The equivalent of the commutation relationship of Λ_+ and Λ_- is given by Kummer (Ref. 2) in his Eq. (3–15). However, he does not further develop angular-momentum-like operators.
[8]See, e.g., R. Pauncz, *Alternate Molecular Orbital Method* (Saunders, Philadelphia and London, 1967), p. 16.
[9]A. J. Coleman, Ref. 3, p. 2; I. Absar and A. J. Coleman, Chem. Phys. Lett. 39, 609 (1976).
[10]A. J. Coleman, remarks at the Reduced Density Matrix Conference, Université de Moncton, Canada, 1977 (unpublished).
[11]The notation is again based on that of Kummer. It is even more important in the case of Γ than in that of L to note that our usage is as defined here and may not be completely equivalent to Kummer's (Ref. 2).
[12]A. J. Coleman, Rev. Mod. Phys. 35, 668 (1963).
[13]D. W. Smith, in *Report of the Density Matrix Seminar*, edited by A. J. Coleman and R. M. Erdahl (Queen's University, Kingston, Ontario, 1968), p. 36.
[14]M. B. Ruskai and J. E. Harriman, Phys. Rev. 169, 101 (1968).
[15]C. Garrod and J. K. Percus, J. Math. Phys. 5, 1756 (1964).
[16]M. Rosina, RDO News No. 2 (Queen's University, Kingston, Ontario, 1976), p. 24.
[17]C. Garrod, M. V. Mikailović, and M. Rosina, J. Math. Phys. 16, 868 (1975).
[18]M. Rosina and C. Garrod, J. Comput. Phys. 18, 300 (1975).
[19]L. J. Kijewski, Phys. Rev. A 9, 2263 (1974).
[20]J. Simons, J. Chem. Phys. 55, 1218 (1971).
[21]J. Simons and J. E. Harriman, Phys. Rev. A 2, 1034 (1970).
[22]This differs from the usual definition of the preimage by a translation by $\underline{X}^{(n)}$. It is more convenient to think of a subset of $\mathfrak{N}_n$ than of $\mathcal{E}_n$.
[23]A. J. Coleman, in *Energy, Structure and Reactivity*, edited by D. W. Smith and W. B. McRae (Wiley, New York, 1973), p. 231.
[24]P. O. Löwdin, Phys. Rev. 97, 1509 (1955).
[25]F. Sasaki and K. Ohno, J. Math. Phys. 4, 1140 (1963).
[26]F. E. Harris, in *Advances in Quantum Chemistry*, Vol. 3, edited by P. O. Löwdin (Academic, New York and London, 1967), p. 61.
[27]V. H. Smith, Jr., J. Chem. Phys. 41, 277 (1964).
[28]R. Pauncz, Ref. 8, Chap. 2.

[29]For an explanation of notation and derivation of results in the spin case, see, e.g., R. Pauncz, Ref. 28.

[30]This differs from the measure introduced by Simons and Harriman (Ref. 21), which dealt with pure-state N representability. The point in $\mathcal{P}^{(n)} - \underline{X}^{(n)}$ closest to $\mathcal{G}_n(D^{(p)})$ will be on the boundary, but need not be a pure state unless it is also on the outer hypersphere defined in I.