# Geometry of density matrices. I. Definitions, $N$ matrices and 1 matrices

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(Received 28 April 1977)

Density matrices defined with respect to a finite basis set are considered as elements in a vector space. A basis set is introduced in the space whose elements are Hermitian matrices. By a suitable transformation of the basis, one component only will contribute to the trace, so the space of unit trace matrices is a translated linear subspace. The positivity constraint is then examined in terms of distance from a multiple of the unit matrix, which is in the interior of the set of density matrices. The boundary of this set is shown to lie between two concentric hyperspheres. The set whose elements are $N$-representable 1 matrices has similar properties.

## I. INTRODUCTION

It is the intent of this paper, and of subsequent papers in this series, to show that potentially useful insights into the nature and properties of density matrices, both statistical and reduced, can be obtained by considering them to be elements in a vector space. In particular, distance relationships are found to be significant. Much of the previous work on the more formal mathematical properties of density matrices has been expressed in topological terms.[1-5] Restriction to finite dimensionality and introduction of a metric allow a more pictorial approach and will supplement other methods. The restriction to finite dimensionality will have consequences less severe than might be imagined if the goal is to relate to practical quantum mechanics, where finite-dimensional approximations are normally necessary. It should also be noted that greater generality than is actually required may unnecessarily obscure meaning for all but the experts.

The ultimate goals of this work involve primarily the elucidation of the theory of reduced density matrices. This paper, however, will be concerned with background and definitions, and with the characterization of the set of $n$-particle or statistical density matrices. Since necessary and sufficient conditions for the $N$-representability of a 1 matrix are known,[1] the set of $N$-representable one-particle reduced density matrices will also be considered. The general process of reduction and relationships involving reduced density matrices will be considered in the next paper.

It seems appropriate to include in Sec. II a review of some of the properties of density matrices. These are all well known, so a brief résumé to establish terminology and notation will suffice. Also included in Sec. II will be the definitions and other preliminaries required to establish a vector space having Hermitian matrices as elements. The subset of density matrices will be considered in Sec. III, and that of $N$-representable 1 matrices in Sec. IV. Conclusions are summarized in Sec. V.

## II. BACKGROUND AND DEFINITIONS

A density matrix, or density operator, $D$ can be defined in any of several equivalent ways.[6-8] One is as a positive, Hermitian operator of unit trace

$$D^\dagger = D, \quad D > 0, \quad \mathrm{tr}D = 1 . \tag{1}$$

We are interested in systems of identical particles and thus require also that the density operator have appropriate symmetry properties. In particular

$$\mathcal{O}D\mathcal{O} = D , \tag{2}$$

where $\mathcal{O}$ is the antisymmetric projection operator (for fermions) or the symmetric projection operator (for bosons). Density matrices may be possible in the statistical sense for other symmetries, but the reduced density matrices which are ultimately of primary interest in the present work would then have no useful role.

Alternatively, if $\{\Psi_k(N)\}$ are a set of orthonormal $n$-particle wave functions of the appropriate symmetry, $\mathcal{O}\Psi_k = \Psi_k$, we can define a density matrix as

$$D(N,N') = \sum_k C_k \Psi_k(N) \Psi_k^*(N') , \tag{3}$$

subject to

$$0 \leq C_k \leq 1, \quad \sum_k C_k = 1 . \tag{4}$$

If $C_k = \delta_{jk}$ so $D = \Psi_j \Psi_j^*$, then

$$[D(N,N')]^2 = \int D(N,N'')D(N'',N')\,dN'' = D(N,N') \tag{5}$$

and conversely. Such a density matrix describes a pure state, while more general $D$'s describe ensembles.

In the general quantum-mechanical situation, the number of possible $\Psi_k$ and the rank of $D$ can be infinite. In actual calculations, however, one deals with finite numbers of functions. In particular, wave functions for many-electron systems are approximated without exception as linear combinations of a finite number of basis functions. We will assume here that a finite set of orthonormal basis functions has been introduced and that all operators are truncated or restricted to the finite-dimensional space spanned by the basis functions. The operators can then be replaced by square matrices of finite dimension. The conditions of Eq. (1) defining a density matrix are then interpreted in the matrix sense.

If a density matrix is defined with respect to functions for $n$ particles, it will be referred to as an $n$ matrix, and will sometimes be indicated $D^{(n)}$ although, when there is no danger of ambiguity, $D$ will be used. We may be interested in properties relating to some smaller number of particles at one time. It will then be useful to introduce a reduced density matrix,[6,9,10] or $p$ matrix, by integrating over some of the variables:

$$D^{(p)}(P,P')=\int D^{(n)}(P,Q;P',Q)\,dQ\,, \tag{6}$$

$P$ being the coordinates of particles $1,\ldots,p$; $Q$ being the coordinates of particles $p+1,\ldots,p+q$; and $N=P,Q$ being the coordinates of all $p+q=n$ particles.

It is readily shown that a reduced density matrix is a density matrix, i.e., it satisfies the conditions (1). However, not every density matrix on $p$-particle space can be obtained from a $D^{(n)}$ by Eq. (6). If this is possible, $D^{(p)}$ is said to be (ensemble) $N$ representable.[1] If at least one $D^{(n)}$ leading to $D^{(p)}$ by Eq. (6) is such that $[D^{(n)}]^2=D^{(n)}$ then $D^{(p)}$ is pure-state $N$ representable and can be associated with an $n$-particle wave function of the appropriate symmetry.

The criteria which must be imposed on a $p$-particle density matrix to assure that it be $N$ representable remain unknown for $p>1$. It is known, however, that necessary and sufficient conditions that $D^{(1)}$ be ensemble $N$ representable are that its eigenvalues $\lambda_k^{(1)}$ be not greater than $1/n$.[1] Other aspects of the $N$-representability problem will be considered in subsequent papers.

Since we are considering here a function space spanned by a finite basis set, we will turn our attention next to finite-dimensional square matrices. Let $\mathcal{S}$ be the set of $R\times R$ Hermitian matrices. Any real linear combination of such matrices is again a member of the set, so $\mathcal{S}$ is a real linear space. We then introduce the usual trace scalar product

$$\begin{aligned}(\underline{U},\underline{V})&=\mathrm{tr}(\underline{U}\underline{V})\\&=\sum_{k,l}U_{kl}V_{lk}=\sum_{k,l}U_{kl}V_{kl}^{*},\quad \underline{U},\underline{V}\in\mathcal{S}\,.\end{aligned} \tag{7}$$

The norm of any element $\underline{U}$ in $\mathcal{S}$ is

$$\|\underline{U}\|=+(\underline{U},\underline{U})^{1/2}=\Big(\sum_{k,l}|U_{kl}|^2\Big)^{1/2}\geq 0\,, \tag{8}$$

and the distance between elements $\underline{U}$ and $\underline{V}$ is

$$d_{\underline{U}\underline{V}}=\|(\underline{U}-\underline{V})\|=\Big(\sum_{k,l}|U_{kl}-V_{kl}|^2\Big)^{1/2}\geq 0. \tag{9}$$

We note that

$$d_{\underline{U}\underline{V}}+d_{\underline{V}\underline{W}}\geq d_{\underline{U}\underline{W}}\geq |d_{\underline{U}\underline{V}}-d_{\underline{V}\underline{W}}|\,. \tag{10}$$

With these definitions $\mathcal{S}$ has become a (Euclidean) vector space of dimension $R^2$.

We next consider a simple basis set or coordinate system for $\mathcal{S}$.[11] The basis elements are themselves Hermitian matrices and are defined by

$$\begin{aligned}&\underline{A}^k\colon\ A^k_{lm}=\delta_{kl}\delta_{lm}\,,\\&\underline{B}^{jk}\colon\ B^{jk}_{lm}=(1/\sqrt{2})(\delta_{jl}\delta_{km}+\delta_{jm}\delta_{kl}),\quad j<k\\&\underline{C}^{jk}\colon\ C^{jk}_{lm}=(i/\sqrt{2})(\delta_{jl}\delta_{km}-\delta_{jm}\delta_{kl}),\quad j<k\,.\end{aligned} \tag{11}$$

There are clearly $R$ distinct $\underline{A}$'s, $\binom{R}{2}$ distinct $\underline{B}$'s, and $\binom{R}{2}$ distinct $\underline{C}$'s. If the $\underline{C}$'s are eliminated from the basis, the result is the $[\frac{1}{2}R(R+1)]$-dimensional space of real, symmetric $R\times R$ matrices.

These basis elements can also be expressed in the "density kernel" form of Eq. (3) in terms of the underlying basis set of functions. The introduction of $R\times R$ matrices assumes a set of orthonormal basis functions. Let these functions be $\{\Phi_k(N)\}$. Then corresponding to the basis matrices defined above we have

$$\begin{aligned}&A^k(N,N')=\Phi_k(N)\Phi_k^{*}(N')\,,\\&B^{jk}(N,N')\\&\quad=(1/\sqrt{2})[\Phi_j(N)\Phi_k^{*}(N')+\Phi_k(N)\Phi_j^{*}(N')],\quad j<k\\&C^{jk}(N,N')\\&\quad=(i/\sqrt{2})[\Phi_j(N)\Phi_k^{*}(N')-\Phi_k(N)\Phi_j^{*}(N')],\quad j<k\,.\end{aligned} \tag{12}$$

We note that the $A$'s are explicitly pure states while the $B$'s and $C$'s are such that

$$(B^{jk})^2=\tfrac{1}{2}(A^j+A^k)=(C^{jk})^2\,. \tag{13}$$

We now return to the matrix form of the basis elements and note that

$$\mathrm{tr}\underline{A}^k=1,\quad \mathrm{tr}\underline{B}^{jk}=\mathrm{tr}\underline{C}^{jk}=0\,. \tag{14}$$

Products of basis elements are simply expressible in terms of the basis elements. From these expressions and the traces, we find that the basis elements are orthonormal.

## III. DENSITY MATRICES

In addition to being Hermitian, a density matrix must be positive and of unit trace. We have seen that each of the $\underline{A}$'s is of trace 1, while the $\underline{B}$'s and $\underline{C}$'s are of trace 0. Now consider an orthogonal transformation of the $\underline{A}$ basis elements among themselves such that one new basis element is

$$\underline{\overline{A}}^0 = (1/\sqrt{R}) \sum_{k=1}^{R} \underline{A}^k . \tag{15}$$

The other new basis elements are given by

$$\underline{\overline{A}}^\kappa = \sum_{k=1}^{R} C_{\kappa k} \underline{A}^k, \quad \kappa = 1, \ldots, R-1 , \tag{16}$$

and the only restriction we need impose at this point on the transformation is that the new set $\{\underline{\overline{A}}^0, \underline{\overline{A}}^\kappa\}$ be orthonormal. Then

$$\mathrm{tr}\underline{\overline{A}}^0 = (1/\sqrt{R})R = \sqrt{R} , \tag{17}$$

$$\mathrm{tr}\underline{\overline{A}}^\kappa = \sum_{k=1}^{R} C_{\kappa k} , \tag{18}$$

but, by assumption,

$$0 = (\underline{\overline{A}}^0, \underline{\overline{A}}^\kappa) = \sum_{k=1}^{R} \sum_{\kappa=1}^{R} \frac{1}{\sqrt{R}} C_{\kappa k} (\underline{A}^l, \underline{A}^k)$$

$$= \frac{1}{\sqrt{R}} \sum_{k=1}^{R} C_{\kappa k} = \frac{1}{\sqrt{R}} \mathrm{tr}\underline{\overline{A}}^\kappa . \tag{19}$$

The set of elements $\underline{\overline{A}}^0, \underline{\overline{A}}^\kappa$, $\kappa = 1, \ldots, R-1$, together with the $\underline{B}$'s and $\underline{C}$'s, provides a complete, orthonormal basis or coordinate system for $\mathcal{E}$. Of these basis elements, only $\underline{\overline{A}}^0$ has nonzero trace.

Let $\mathfrak{N}$ be the space spanned by the $R^2 - 1$ basis elements of $\mathcal{E}$ other than $\underline{\overline{A}}^0$. Any element in $\mathcal{E}$ may be thought of as a multiple of $\underline{\overline{A}}^0$ combined with an element of $\mathfrak{N}$. In particular, any unit-trace element of $\mathcal{E}$ can be expressed as

$$\underline{V}' = \underline{V} + \underline{X} , \tag{20}$$

where $\underline{V}$ has $\underline{\overline{A}}^0$ component 0 and corresponds uniquely to an element of $\mathfrak{N}$, while

$$\underline{X} = R^{-1/2} \underline{\overline{A}}^0 = R^{-1} \sum_{k=1}^{R} \underline{A}^k . \tag{21}$$

The special element $\underline{X}$ is a multiple of the unit matrix in $\mathcal{E}$ and corresponds to the origin of $\mathfrak{N}$. We can think of density matrices, along with other unit-trace Hermitian matrices, as elements of $\mathcal{E}$ or in terms of the corresponding elements of $\mathfrak{N}$.

We turn now to an investigation of positivity, the remaining condition characterizing a density matrix. We initially define a set of matrices

$$\underline{T}(t) = t^{-1} \underline{Q}(t), \quad t = 1, 2, \ldots, R \tag{22}$$

where $\underline{Q}(t)$ is an idempotent matrix having $t$ eigenvalues equal to 1 and the remaining $R - t$ eigenvalues equal to 0. Since $\mathrm{tr}[\underline{T}(t)] = 1$ and $T(t)$ has no negative eigenvalues, it is a density matrix. We note that $\underline{T}(R) = \underline{X}$ and will use the special symbol $\underline{P}$ for $\underline{T}(1)$, since $\underline{T}(1) = \underline{Q}(1)$ is a pure-state density matrix.

Since $\underline{X}$ is a multiple of the unit matrix, its product with any element of $\mathcal{E}$ is

$$\underline{X}\underline{V} = \underline{V}\underline{X} = R^{-1} \underline{V} . \tag{23}$$

Using these results, we find the distance between $\underline{T}(t)$ and $\underline{X}$ to be

$$d_{\underline{T}\underline{X}} = \{\mathrm{tr}[(\underline{X} - \underline{T})^2]\}^{1/2}$$

$$= \{\mathrm{tr}[(1/R)\underline{X} - (2/R)\underline{T} + (t^{-1}\underline{Q})^2]\}^{1/2}$$

$$= \left(\frac{1}{R} - \frac{2}{R} + \frac{1}{t}\right)^{1/2} = \left(\frac{R-t}{Rt}\right)^{1/2} . \tag{24}$$

In particular, when $t = 1$ we find that any pure-state density matrix is at a distance of $[(R-1)/R]^{1/2}$ from $\underline{X}$ or, equivalently, *any element of* $\mathfrak{N}$ *corresponding to a pure-state density matrix lies on a hypersphere of radius* $[(R-1)/R]^{1/2}$ *about the origin.*

Traces, and therefore distances in $\mathcal{E}$ or $\mathfrak{N}$, are independent of any unitary transformation of all the matrices involved. We can think of parametrizing an element in $\mathcal{E}$ not by the $R^2$ real numbers that are its components in the coordinate system of $\mathcal{E}$, but by $R$ eigenvalues and $R(R-1)$ other real numbers which determine the transformation diagonalizing that particular matrix. (These parameters may not all be independent if some of the eigenvalues are degenerate.) The element $\underline{X}$ is invariant under unitary transformations. In computing the distance of some other element $\underline{V}$ from $\underline{X}$ we can choose the basis in which $\underline{V}$ is diagonal, and we find that

$$d_{\underline{V}\underline{X}} = \left[\sum_{j=1}^{R} \left(\frac{1}{R} - \lambda_j\right)^2\right]^{1/2} , \tag{25}$$

where the $\lambda_j$ are the eigenvalues of $\underline{V}$.

Suppose that $\underline{D}$ is a density matrix having more than one nonzero eigenvalue (an ensemble density matrix). Since no eigenvalue can be negative and they must sum to 1, each must be less than 1. This means that

$$\lambda_k^2 < \lambda_k \tag{26}$$

for those eigenvalues which are nonzero, and

$$d_{\underline{D}\underline{X}}=\left[\sum_{j=1}^{R}\left(\frac{1}{R}-\lambda_j\right)^2\right]^{1/2}$$
$$=\left[\sum_{j=1}^{R}\left(\frac{1}{R^2}-\frac{2\lambda_j}{R}+\lambda_j^2\right)\right]^{1/2}$$
$$<\left[\sum_{j=1}^{R}\left(\frac{1}{R^2}-\frac{2\lambda_j}{R}+\lambda_j\right)\right]^{1/2}=\left(\frac{1}{R}-\frac{2}{R}+R\right)^{1/2}$$
$$=\left(\frac{R-1}{R}\right)^{1/2}. \tag{27}$$

*Any ensemble density matrix lies inside the hypersphere on which the pure-state density matrices lie.*[12] We note further that if $\underline{D}$ is a density matrix lying on the hypersphere

$$d^2_{\underline{D}\underline{X}}=\left(\frac{R-1}{R}\right)=\sum_{j=1}^{R}\left(\frac{1}{R}-\lambda_j\right)^2=\frac{1}{R}-\frac{2}{R}+\sum_{j=1}^{R}\lambda_j^2,$$
$$\sum_{j=1}^{R}\lambda_j^2=1=\sum_{j=1}^{R}\lambda_j. \tag{28}$$

This can occur for nonnegative $\lambda_j$ only if one of them is 1 and the rest are 0, i.e., $\underline{D}$ is a pure state. *Any density matrix on the hypersphere is a pure-state density matrix.* Not all elements on the hypersphere are density matrices, however. Counterexamples are readily found, except in the special case $R=2$.

We now seek to further characterize the subspace of $\mathfrak{N}$ whose elements correspond to density matrices. We begin by proving a theorem which will also be useful in the next section.[13]

*Theorem.* Let $\{x_i, i=1,\ldots,m\}$ be a set of real numbers subject to the sum constraint $\sum_{i=1}^{m}x_i=1$, and let $\sigma=\sum_{i=1}^{m}x_i^2$. Then (a) $\sigma\geq 1/m$, with equality if and only if $x_i=1/m$, all $i$. If the $x_i$ are further subject to the constraint $1\leq x_i\leq 1/k$, where $k$ is an integer $\leq m$, then (b) $\sigma\leq 1/k$, with equality if and only if $k$ of the $x_i$ are equal to $1/k$ and the $m-k$ remaining $x_i$ are zero.

The proof of (a) is straightforward. Consider

$$\sum_{i=1}^{m}\left(x_i-\frac{1}{m}\right)^2=\sigma-\frac{1}{m}. \tag{29}$$

Then

$$\sigma=\frac{1}{m}+\sum_{i=1}^{m}\left(x_i-\frac{1}{m}\right)^2\geq\frac{1}{m} \tag{30}$$

with equality only if each term in the sum is zero.

To prove (b) we suppose that $\{x_i\}$ is any set satisfying the sum and range conditions. A sequence of sets $\{x_i^{(n)}\}$ each having its associated $\sigma^{(n)}$, are then defined beginning with $x_i^{(0)}=x_i$, all $i$, and proceeding iteratively as follows: If each $x_i^{(n)}$ is either 0 or $1/k$, then $\sigma^{(n)}=1/k$ and the iterative cycle is finished. If, however, there are $x_i^{(n)}$ in the open interval $(0,1/k)$, let $x_a^{(n)}$ be the smallest and $x_b^{(n)}$ the largest of this subset. Now let $\Delta=\min(x_a^{(n)},(1/k)-x_b^{(n)})$, and define a new set with

$$x_i^{(n+1)}=\begin{cases}x_a^{(n)}-\Delta & \text{if } i=a,\\ x_b^{(n)}+\Delta & \text{if } i=b,\\ x_i^{(n)} & \text{if } i\neq a,b.\end{cases} \tag{31}$$

Then either $x_a^{(n+1)}=0$ or $x_b^{(n+1)}=1/k$ and the $x_i^{(n+1)}$ satisfy both sum and range constraints, ready for the next cycle of iteration. Now,

$$\sigma^{(n+1)}=(x_a^{(n)}-\Delta)^2+(x_b^{(n)}+\Delta)^2+\sum_{i\neq a,b}(x_i^{(n)})^2$$
$$=\sigma^{(n)}+2(x_a^{(n)}+x_b^{(n)})\Delta+\Delta^2>\sigma^{(n)}. \tag{32}$$

This suffices to establish the result.

It may be noted that case (b), with $x_i=\lambda_i$, $m=r$, and $k=1$ provides an alternative proof of an earlier result. We now consider the question of how closely a nondensity matrix can approach to $\underline{X}$. A Hermitian matrix of unit trace which is not a density matrix must have at least one negative eigenvalue. Suppose that $\underline{V}$ is such a matrix, and that, of its eigenvalues $\lambda_i$ $q$ are negative while $p=R-q$ are non-negative. Let the sum of the nonnegative eigenvalues be $p\alpha$ and that of the negative eigenvalues be $q\beta$, and define $x_i=\lambda_i/p\alpha$ when $\lambda_i\geq 0$ or $y_i=\lambda_i/q\beta$ when $\lambda_i<0$. Then $\sum x_i=\sum y_i=1$ and

$$d^2_{\underline{V}\underline{X}}=\sum_{i=1}^{R}\left(\lambda_i-\frac{1}{R}\right)^2=\sum_{i=1}^{R}\lambda_i^2-\frac{1}{R}$$
$$=(p\alpha)^2\sum x_i^2+(q\beta)^2\sum y_i^2-\frac{1}{R}$$
$$\geq p\alpha^2+q\beta^2-\frac{1}{R}=\frac{1}{R-q}(1-2q\beta+R\beta^2)-\frac{1}{R}, \tag{33}$$

where the inequality is obtained by application of part (a) of the theorem to the set of $p$ $x_i$'s and to the set of $q$ $y_i$'s individually. From its definition, $\beta\leq 0$ so this lower bound will be minimized if the magnitude of $\beta$ is made as small as possible. For any $\underline{V}$ with $q$ eigenvalues at least slightly less than zero, then, $d^2_{\underline{V}\underline{X}}\geq(R-q)^{-1}-R^{-1}=d^2_{\underline{X}\underline{T}(p)}$. The closest approach of such a $\underline{V}$ to $\underline{X}$ will occur for $q=1$ and $\beta\simeq 0_-$, in which case $\underline{V}$ is essentially $\underline{T}(R-1)$. We conclude that *there is a second hypersphere centered at the origin in* $\mathfrak{N}$ *(or centered at* $\underline{X}$ *in* $\mathcal{E}$*), of radius*

$$d_{\underline{X}\underline{T}(R-1)}=\left[(R-1)\left(\frac{1}{R}-\frac{1}{R-1}\right)^2+\frac{1}{R^2}\right]^{1/2}$$
$$=[R(R-1)]^{-1/2}, \tag{34}$$

*with the property that any element in $\mathfrak{N}$ inside this hypersphere corresponds to a density matrix.*

Any density matrix having one or more zero eigenvalues must lie on the boundary of the space of density matrices. In particular, if $\underline{D}$ is a density matrix with eigenvalue $i$ equal to 0, let $\underline{V}$ be a matrix with the same eigenvector but with eigenvalue $i$ equal to $-\delta$ and each other eigenvalue $\delta/(R-1)$ greater than the corresponding eigenvalue of $\underline{D}$. Then $\mathrm{tr}(\underline{V})=1$, but $\underline{V}$ is not a density matrix (for $\delta>0$) and $d_{\underline{D}\underline{V}}=[R/(R-1)]^{1/2}\delta$ can be made as small as desired by taking $\delta$ sufficiently small.

Let $c$ be the eigenvector associated with eigenvalue 1 for a pure-state density matrix $\underline{P}$. Then, in terms of components with respect to the original orthonormal basis set $\{\Phi_k\}$,

$$P_{jk}=c_j c_k^* \tag{35}$$

and for any positive matrix $\underline{Q}$,

$$(\underline{P},\underline{Q})=\sum_{j,k}P_{kj}Q_{jk}=\sum_{j,k}c_j^*Q_{jk}c_k\geq 0\,. \tag{36}$$

The positivity condition is thus equivalent to the requirement that the scalar product of a positive matrix with any pure state is positive (or zero). If $\{\underline{D}^{(i)}\}$ is a set of density matrices and

$$\underline{V}=\sum_j a_j\underline{D}^{(j)}\,, \tag{37}$$

then $\mathrm{tr}(\underline{V})=1$ and

$$(\underline{P},\underline{V})=\sum_j a_j(\underline{P},\underline{D}^{(j)})\,. \tag{38}$$

If the $a_i$ are all positive and sum to 1, $\underline{V}$ will be positive and of trace 1, thus a density matrix. This is the well-known *convexity* property of the set of density matrices.[1]

The set whose elements are density matrices may be thought of as a subset of $\mathfrak{N}$ having the same dimension, $R^2-1$, as $\mathfrak{N}$. We seek to further characterize the boundary of this subset. Let $\mathcal{P}$ be the subset and $\partial\mathcal{P}$ its boundary. The next property to be shown is that $\partial\mathcal{P}$ is a closed continuous hypersurface of dimension $R^2-2$. We know that $\underline{X}$, the origin of $\mathfrak{N}$, is in $\mathcal{P}$. Consider now a ray of elements of $\mathfrak{N}$, $\alpha\underline{Q}$, with $\alpha>0$ and the corresponding set of elements in $\mathcal{S}$,

$$\underline{Q}'(\alpha)=\underline{X}+\alpha\underline{Q}\,. \tag{39}$$

The matrix $\underline{Q}$ must have trace 0, and it cannot be the zero matrix. It must thus have at least one negative eigenvalue. Let $q_-$ be the magnitude of the most negative eigenvalue of $\underline{Q}$. Then if $\alpha q_-<1/R$, $Q'(\alpha)>0$, but if $\alpha>1/Rq_-$, $\underline{Q}'(\alpha)$ will have at least one negative eigenvalue. We conclude that the ray $\alpha\underline{Q}$ intersects $\partial\mathcal{P}$ at the point $(1/Rq_-)\underline{Q}$ [as an element of $\mathcal{S}$ the point is $Q'(1/R_{q_-})$]. This ray cannot have another intersection with $\partial\mathcal{P}$ because of the convexity of $\mathcal{P}$. Any ray from the origin of $\mathfrak{N}$ can be expressed in this way for some appropriate choice of $\underline{Q}$. Each element in $\partial\mathcal{P}$ satisfies the one additional condition determining $\alpha$, so $\partial\mathcal{P}$ is of dimension $R^2-2$, one less than that of $\mathfrak{N}$. If it can be established that $\partial\mathcal{P}$ is continuous, the desired result will have been established.

In order for $\partial\mathcal{P}$ to be discontinuous, in the sense that the term continuity is intended here, it would be necessary that two rays arbitrarily close together intersect $\partial\mathcal{P}$ at finitely different distances from $\underline{X}$. It can be shown that this is impossible. Let $\underline{Q}$ and $\underline{S}$ be elements of $\mathfrak{N}$, scaled if necessary so that $||\underline{Q}||=||\underline{S}||=1$, and consider elements of $\mathcal{S}$

$$\begin{aligned}\underline{Q}'(\alpha)&=\underline{X}+\alpha\underline{Q}\,,\\ \underline{S}'(\beta)&=\underline{X}+\beta\underline{S}\,.\end{aligned} \tag{40}$$

The intersections of $\underline{Q}'(\alpha)$ and $\underline{S}'(\alpha)$ with $\partial\mathcal{P}$ occur at distances from $\underline{X}$ of $1/(q_-R)$ and $1/(s_-R)$, respectively, where $q_-$ is the magnitude of the most negative eigenvalue of $\underline{Q}$ and $s_-$ is the magnitude of the most negative eigenvalue of $\underline{S}$. If these distances were to be finitely different for rays $\alpha\underline{Q}$ and $\beta\underline{S}$ arbitrarily close together it would be necessary that matrices with elements differing by arbitrarily small amounts have finitely different eigenvalues. This is impossible,[14] so $\partial\mathcal{P}$ is continuous.

The hypersurface $\partial\mathcal{P}$ must lie between the two hyperspheres considered previously, and the points where it intersects the outer hypersphere are the pure-state density matrices. Let $\underline{P}=\underline{X}+\hat{\underline{P}}$ be a pure state and consider $\underline{Q}'(\alpha)=\underline{X}-\alpha\hat{\underline{P}}$. In the basis where $\underline{P}$ is diagonal, $\hat{\underline{P}}$ and $\underline{Q}'(\alpha)$ will also be diagonal. One of the eigenvalues of $\hat{\underline{P}}$ is $1-1/R$ and the rest are $-1/R$. It follows that $\underline{Q}'(\alpha)$ has one eigenvalue

$$q_i=1/R-\alpha(1-1/R)=(1+\alpha)/R-\alpha \tag{41}$$

while the rest are

$$q_j=(1+\alpha)/R\quad\text{all } j\neq i\,. \tag{42}$$

For $\alpha\geq 0$, only $q_i$ can possibly be negative. It will be so if

$$\alpha>(1+\alpha)/R,\quad \alpha>1/(R-1)\,. \tag{43}$$

The point at which this eigenvalue is zero lies in $\partial\mathcal{P}$. It will be denoted by $\tilde{\underline{Q}}$:

$$\tilde{\underline{Q}}=\underline{Q}'\left(\alpha=\frac{1}{R-1}\right)=\underline{X}-\frac{1}{R-1}\,\hat{\underline{P}}\,. \tag{44}$$

Its distance from $\underline{X}$ is

$$d_{\tilde{Q}\underline{X}} = \frac{1}{R-1}\left|\left|\hat{\underline{P}}\right|\right| = \frac{1}{R-1} d_{\underline{P}\underline{X}} = \frac{1}{R-1}\left(\frac{R-1}{R}\right)^{1/2}$$
$$= [R(R-1)]^{-1/2}. \tag{45}$$

The conclusion is that *immediately opposite (in the sense of inversion through the origin or $\underline{X}$) any point where $\partial\mathcal{P}$ intersects the outer hypersphere is a point where it intersects the inner hypersphere.*

Before leaving this topic, we note a few local properties of $\partial\mathcal{P}$. Any two elements of $\mathcal{S}$ with the same eigenvalues are either both in the interior of $\mathcal{P}$, both outside $\mathcal{P}$, or both in $\partial\mathcal{P}$. They are also at the same distance from $\underline{X}$. For any element of $\mathcal{S}$ other than a multiple of the unit matrix there are other, distinct elements having the same eigenvalues, including some which are arbitrarily close to the original element. It follows that at any point in the surface $\partial\mathcal{P}$ there are some directions in which $\partial\mathcal{P}$ is curved and in fact contains a circle centered at $\underline{X}$. This means that $\partial\mathcal{P}$ can have no "points".

On the other hand, $\partial\mathcal{P}$ contains linear subsets. For example, a linear combination of two diagonal matrices which have a zero diagonal element in the same place will also have a zero diagonal element (eigenvalue) in that place and, if all other eigenvalues are non-negative, will be in $\partial\mathcal{P}$.

### Another characterization of positive matrices

There is of course another way in which the set of positive Hermitian matrices can be characterized: let $\underline{D} = \underline{V}^2$, where $\underline{V}$ is an arbitrary element of $\mathcal{S}$. Then $\underline{D}$ will be positive. The requirement that $\mathrm{tr}(D) = 1$ is equivalent to the requirement that $\left|\left|\underline{V}\right|\right| = 1$. A mapping is thus established of the unit hypersphere in $\mathcal{S}$ onto $\mathcal{P}$.[2] Unfortunately, it is nonlinear and many to one. If $\underline{D}$ has $t$ nonzero eigenvalues, there will be $2^t$ distinct $\underline{V}$'s with different choices for signs for their eigenvalues, which all map into $\underline{D}$. The many-to-one aspect would be an irritation but not an insurmountable obstacle. It is the nonlinearity which makes this approach of little value for us.

Let us consider not the general case but the much simpler case of a pure-state density matrix $\underline{P}$. It will be such that $\mathrm{tr}(\underline{P}) = 1$ and $\underline{P}^2 = \underline{P}$. These two conditions suffice to establish positivity. The latter is a special case of $\underline{D} = \underline{V}^2$, with $\underline{D} = \underline{V}$, and the pure states are seen to lie in the intersection of the unit sphere in $\mathcal{S}$ with $\mathcal{N}$, which is just the outer hypersphere obtained previously. The equation $\underline{P}^2 = \underline{P}$ provides a set of simultaneous quadratic equations in the matrix elements, or the expansion coefficients in the $\underline{A},\underline{B},\underline{C}$ basis, of $\underline{P}$. Conditions for the existence of a solution of a set of simultaneous quadratic equations are very difficult, in general. (Were this not the case, the $N$-representability problem would have been solved long ago.) In the present situation, however, a straightforward solution is readily available: $\underline{P}$ will be such that $\mathrm{tr}(\underline{P}) = 1$ and $\underline{P}^2 = \underline{P}$ if and only if $\underline{P}_{jk} = c_j c_k^*$, where the $c_j$ are a set of $R$ complex numbers such that $\sum_j |c_j|^2 = 1$. Of course $c$ is just the eigenvector of $\underline{P}$ associated with the eigenvalue 1, so once again we have found that the wave function provides the best parametrization of a pure-state density matrix.

## IV. $N$-REPRESENTABLE 1 MATRICES

We turn to the case in which the underlying functions $\Psi_k$ and $\Phi_k$ are one-electron functions, or spin orbitals. Let $\mathcal{P}^{(1)}$ be the set of density matrices in this case and let $\mathcal{P}_N^{(1)}$ be the subset of $\mathcal{P}^{(1)}$ whose elements are (ensemble) $N$-representable reduced density matrices. A Hermitian matrix will be in $\mathcal{P}_N^{(1)}$ if and only if its eigenvalues are such that

$$0 \le \lambda_j \le 1/n, \quad j = 1, \ldots, R$$
$$\sum_{j=1}^{R} \lambda_j = 1. \tag{46}$$

It is clearly necessary that $R \ge n$, and we note that $\underline{X}$ is in $\mathcal{P}_N^{(1)}$. Part (b) of the theorem proved earlier then immediately gives, for any $N$-representable 1 matrix $\underline{V}$,

$$d_{\underline{V}\underline{X}}^2 = \sum_{i=1}^{R} \lambda_i^2 - \frac{1}{R} \le 1/n - 1/R = (R-n)/Rn. \tag{47}$$

Equality occurs when $\underline{V} = \underline{T}(n)$. Such a matrix is, of course, the pure-state reduced density matrix for a Hartree-Fock function (i.e., a single Slater determinant). We have shown that these Slater 1 matrices lie on a hypersphere of radius $[(R-n)/Rn]^{1/2}$ about $\underline{X}$, and that any other $N$-representable 1 matrix, ensemble, or non-Slater pure state, will be inside this hypersphere. It is also readily seen that any $N$-representable 1 matrix on the hypersphere will be a Slater 1 matrix.[15]

A Hermitian matrix of unit trace can fail to be an $N$-representable 1 matrix by having one or more eigenvalues which are negative or greater than $1/n$. No assumption about an upper limit on the size of an eigenvalue was made in the analysis associated with Eq. (45), so it still applies. If we define new parameters $\mu_i$ by setting

$$\lambda_i = 1/n - \mu_i, \tag{48}$$

then $\lambda_i$ will be greater than $1/n$ if $\mu_i$ is negative. It is found that for $\underline{V}$ with eigenvalues $\lambda_i$,

$$d^2_{\underline{Y}\underline{X}} = \sum_i \left(\frac{1}{n} - \mu_i - \frac{1}{R}\right)^2$$
$$= \sum_i \mu_i^2 - \frac{(R-n)^2}{Rn^2} \geq p\alpha^2 + q\beta^2 - \frac{1}{R}\xi^2$$
$$= [1/(R-q)](\xi^2 - 2q\xi\beta + R\beta^2) - (1/R)\xi^2, \quad (49)$$

where now $p,q$ are the numbers of $\mu_i$'s which are non-negative or negative and $\alpha,\beta$ are their averages, respectively, and $\xi = (R-n)/n$. As in the case of negative eigenvalues, the closest approach to $\underline{X}$ occurs when $\beta \simeq 0_+$ and $q = 1$, and is

$$d^2_{\underline{Y}\underline{X}} = \left(\frac{1}{R-1} - \frac{1}{R}\right)\xi^2 = \frac{1}{R(R-1)}\frac{(R-n)^2}{n^2}. \quad (50)$$

An inner hypersphere can be defined in this case with radius

$$d_{\min} = \begin{cases} [R(R-1)]^{-1/2} & \text{if } n \leq \frac{1}{2}R \\ [(R-n)/n][R(R-1)]^{-1/2} & \text{if } n > \frac{1}{2}R. \end{cases} \quad (51)$$

Any unit-trace element of $\mathcal{E}$ inside a hypersphere of radius $d_{\min}$ about $\underline{X}$ (corresponding to an element of $\mathfrak{N}$ within a hypersphere of radius $d_{\min}$ about the origin of $\mathfrak{N}$) will be an $N$-representable 1 matrix. The boundary $\partial\mathcal{P}_N^{(1)}$ of $\mathcal{P}_N^{(1)}$ will lie between this inner hypersphere and the outer hypersphere of radius $[(R-n)/Rn]^{1/2}$.

The remarks in the previous section about curvature and linear subsets within the surface apply to $\partial\mathcal{P}_N^{(1)}$ as well. The discussion of continuity can also readily be extended to apply in this case, but it is not true that opposite a point where $\partial\mathcal{P}_N^{(1)}$ intersects the outer hypersphere is a place where it intersects the inner hypersphere.

## V. CONCLUSIONS

It has been shown in this paper that density matrices defined with respect to a finite basis set can be thought of as elements in a vector space. Not all elements in this space, the space of Hermitian operators of trace 1, are density matrices because not all of them are positive. There are two concentric hyperspheres in the space with the property that the bounding hypersurface of the subset whose elements are density matrices lies between them. An element in the intersection of the boundary with the outer hypersphere is a pure-state density matrix, and vice versa. Immediately opposite the point where the surface intersects the outer hypersphere is a point where it intersects the inner hypersphere. The boundary contains linear subsets, but at any point on the boundary there are some directions in which it is curved.

All of these results serve to give a somewhat better picture of the set of density matrices. Unfortunately, they do not provide a complete characterization of this set. The characterization in terms of a nonlinear mapping is straightforward, but not really very convenient. It will be shown in the following paper that a sufficiently convenient characterization of the set of density matrices would provide a solution to the $N$-representability problem.

Since the ensemble $N$-representability problem for the 1 matrix has been solved, the set whose elements are $N$-representable density matrices can also be investigated. It is, of course, contained within the set of density matrices. Its boundary also lies between two hyperspheres, intersecting the outer one in 1 matrices corresponding to Slater determinants. The inner hypersphere coincides with the inner hypersphere for the density matrices if $n$ is less than or equal to half the number of basis functions, and is inside that hypersphere for larger $n$. The relationship between this set for one-particle density matrices and the set of $n$-particle density matrices will be considered in the following paper.

## ACKNOWLEDGMENT

This work was supported by the National Science Foundation under Grant No. CHE74-17494-A01.

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[11] Cf. Fano, Ref. 8, Sec. 6.
[12] It is well known that the pure states are extreme in the space of density matrices. Cf., e.g., Refs. 1 and 2.

[13]No claim of originality is made for these results, but it is easier to give a proof than to locate one and relate it to the present circumstances.

[14]J. H. Wilkinson, *The Algebraic Eigenvalue Problem* (Clarendon Press, Oxford, 1965), p. 63, quoting A. M. Ostrowski [J. Ber. Deutsch. Math. Verein 60, 40–42 (1957)].

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