CONDITIONS FOR THE REPRESENTABILITY OF THE DENSITY MATRIX

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The necessary and sufficient condition for the N-representability of reduced density matrices is found for functions of limited rank.

1. The so-called N-representability problem is lately attracting considerable attention [1-5]. We recall that it consists of developing the necessary and sufficient conditions for which some function $P(x_1, x_2. \ldots, x_s|x_1', x_2', \ldots, x_s')$, antisymmetric in all groups of arguments (the argument represents the set of all the one-particle spatial and spin coordinates), Hermitian, and which is a kernel of a positive definite nuclear operator, can be considered an s-th-order reduced density matrix $P_s^{(N)}(x|x')$ of a pure or mixed state of a system of N particles. The lower index s (which for wavefunctions will be enclosed in parentheses in the following) indicates the number of arguments, which for simplicity of notation are denoted by the letter x.

The solution of this problem for a second-order reduced density matrix of a system of particles interacting in pairs in conjunction with the variational principle would allow essentially to remove the many-particle wavefunction $\Psi_{(N)}(x)$ from the treatment of the majority of quantum-mechanical problems.

At the present time the N-representability problem is still far from a constructive solution. Its discussion is usually conducted in terms of constraints imposed by the requirement of N-representability regarding the spectral decomposition of $P_s^{(N)}(x|x')$:

$$P_s^{(N)}(x|x') = \sum_{i=1} \Lambda_i^{(s)} \varphi_{(s)i}(x) \varphi_{(s)i}^*(x'). \tag{1}$$

The following terminology is widely used [1, 2]. The eigenfunctions $\varphi_{(s)i}(x)$ (corresponding to $\Lambda_j^{(s)} \neq 0$) of an s-th-order reduced density matrix are called natural s-particle states (in an N-particle system), and the eigenvalues $\Lambda_i^{(s)}$ are called natural occupation numbers of the corresponding states. The number of terms in the expansion (1) with nonzero $\Lambda_i^{(s)}$ is usually designated the s-th rank of the wavefunction $\Psi_{(N)}(x)$, and the first rank is simply rank. The function $\Psi_{(N)}(x)$ itself, for which $P_s^{(N)}(x|x')$ is an s-th-order reduced density matrix

$$P_s^{(N)}(x|x') = \binom{N}{s} \int_y \Psi_{(N)}(xy) \Psi_{(N)}^*(x'y), \tag{2}$$

we shall call the representation function.

For a mixed-state representation the N-representability problem was solved in the case of $P_1^{(N)}(x|x')$ [1], and some progress has been made for $P_2^{(N)}(x|x')$ [3].

For a pure-state representation the complete solution exists only for the ranks r = N + 1 and r = N + 2 [1, 2] for a first-order reduced density matrix, and also for a second-order reduced density matrix in the case of three particles [4]. A general condition of N-representability of a first-order reduced density matrix for a system of many particles described by a Hamiltonian invariant under time reversal was found in [5], and was generalized to the degenerate case in [6]. Several results pertaining to the representation

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by functions of a special type were obtained in [7] for a many-particle system, and they were extended also to systems with a small number of particles in [8].

The necessary and sufficient condition for the N-representability of a reduced density matrix of arbitrary s for functions of limited rank $\Psi_{(N)}(x)$ is formulated in this paper.

According to the Carlson–Keller theory [2, 9] a function of rank r can be represented in the form

$$\Psi_{(N)}(x) = \sum_{i=1}^{r} \varphi_i(x_1)\,\theta_i(x_2, x_3, \ldots, x_N), \tag{3}$$

where $\varphi_1(x_1)$ are natural one-particle states, and $\theta_i(x_2, x_3, \ldots, x_N)$ are natural $(N-1)$-particle states. It can be shown that if an antisymmetric function $\Psi_{(N)}(x)$ is represented in the form of Eq. (3) then it is a superposition of $\binom{r}{N}$ determinants constructed from one-particle functions $\varphi_{i_1}, \varphi_{i_2}, \ldots, \varphi_{i_N}$ taken from the sequence $\varphi_1, \varphi_2, \ldots, \varphi_r$ [10]:

$$\Psi_N(x) = \sum_{\{i\}_N} c_{i_1 i_2 \ldots i_N} \det_N[\varphi_{i_1}, \varphi_{i_2}, \ldots, \varphi_{i_N}](x). \tag{4}$$

Consider a "large" determinant constructed from all the basis functions

$$O(x_1, x_2, \ldots, x_r) = \det_r[\varphi_1, \varphi_2, \ldots, \varphi_r](x) \equiv \frac{1}{\sqrt{r!}} \begin{vmatrix} \varphi_1(x_1)\,\varphi_1(x_2) \ldots \varphi_1(x_r) \\ \varphi_2(x_1)\,\varphi_2(x_2) \ldots \varphi_2(x_r) \\ \cdots\cdots\cdots\cdots \\ \varphi_r(x_1)\,\varphi_r(x_2) \ldots \varphi_r(x_r) \end{vmatrix}. \tag{5}$$

If by $Q_N^{(r)}$ we denote the N-th-order reduced density matrix for this determinant then

$$\int_{x'} Q_N^{(r)}(x|x')\,\Psi_{(N)}(x') = \Psi_{(N)}(x), \tag{6}$$

for, as is evident from Eq. (4), $\Psi_N(r)$ is the natural N-state for O. In that case according to the Carlson–Keller theory there exists a natural r – N = M-state $\Phi_{(M)}(y)$, also belonging to 1:

$$\Phi_{(M)}(y) = \sqrt{\binom{r}{N}} \int_x \Psi^*_{(N)}(x)\,O(xy) = \sum_{\{i\}_N} (-1)^{\alpha\{i\}} c^*_{i_1 i_2 \ldots i_N} \det_M[\varphi_{j_1}, \varphi_{j_2}, \ldots, \varphi_{j_M}](y), \tag{7}$$

where $\alpha\{i\} = 1 + 2 + \ldots + N + i_1 + i_2 + \ldots + i_N$. The multiplier $\sqrt{\binom{r}{N}}$ normalizes $\Phi_{(M)}(y)$ to unity. In evaluating the right side of Eq. (7) we have used the rule for integrating an alternating product and expanded O(xy) by the Laplace formula through the first N columns. $j_1, j_2, \ldots, j_M$ together with $i_1, i_2, \ldots, i_N$ form a complete set of indices 1, 2, ..., r. We shall call the functions $\Psi_{(N)}(x)$ and $\Phi_{(M)}(x)$ reciprocal. It it easy to show that

$$\Psi_{(N)}(x) = \sqrt{\binom{r}{M}} \int_y \Phi^*_{(M)}(y)\,O(xy). \tag{8}$$

If $\Psi_{(N)}$ describes a system of N particles, $\Phi_{(M)}$ describes a system of M holes. A comparison of Eqs. (7) and (4) reveals the reason why with a limited rank for every wavefunction $\Psi_{(N)}$ it is possible to construct a reciprocal function $\Phi_{(M)}$. If the rank is limited, then the coefficients in the expression of the type (4) can be counted both as being present in the determinant and as an additional set of indices, which together with the indices of the one-electron functions of the determinant form the complete set of indices 1, 2, ..., r. Therefore, the same set of coefficients can serve as a basis for the construction of the function of N and r – N particles.

We shall now find the relationship between the reduced density matrices $P_s^{(N)}(x|x')$ and $P_s^{(M)}(x|x')$ of the functions $\Psi_{(N)}(x)$ and $\Phi_{(M)}(y)$. By definition of the reduced density matrix (2) and using Eq. (8)

$$P_s^{(N)}(x|x') = \binom{N}{s} \int_u \Psi_{(N)}(xu)\,\Psi^*_{(N)}(x'u) = \binom{M+s}{s} \int_{y,z} \Phi^*_{(M)}(y)\,Q^{(r)}_{M+s}(xy|x'z)\,\Phi_{(M)}(z). \tag{9}$$

We use for the determinant in the reduced density matrix the expression

$$Q^{(r)}_{M+s}(xy|x'z) = A_{xy}[p(x_1|x_1'),\ldots,p(x_s|x_s'),p(y_1|z_1),\ldots,p(y_M|z_M)]$$
$$= A_{xy}[Q^{(r)}_s(x|x'),p(y_1|z_1),\ldots,p(y_M|z_M)], \tag{10}$$

where A_{XY} is an antisymmetrizing operator in all x- and y-variables, and

$$p(x_1|x_1') \equiv Q^{(r)}_1(x_1|x_1') = \sum_{i=1}^{r} \varphi_i(x_1)\varphi_i^*(x_1'). \tag{10'}$$

Since according to Eq. (7) $\Phi_{(M)}(y)$ is separable in each variable only into the one-electron functions which appear in $p(x_1|x_1')$, it is not changed by the action $p(y_1|z_1),\ldots,p(y_M|z_M)$, and

$$\int_z Q^{(r)}_{M+s}(xy|x'z)\Phi_{(M)}(z) = A_{xy}[Q^{(r)}_s(x|x')\Phi_{(M)}(y)]. \tag{11}$$

Substituting this expression into Eq. (9), using the Sasaki formula [1] for A_{XY} and the Hermitian property of the operator A_y, and taking account of the antisymmetry of the corresponding functions, we have (k = min (M, s))

$$P^{(N)}_s(x|x') = A_x\sum_{j=1}^{k}(-1)^j\binom{s}{j}\binom{M}{j}\int_y \Phi^*_{(M)}(y)\,\Phi_{(M)}(x_1,\ldots,x_j,y_{j+1},\ldots,y_M)\,Q^{(r)}_s(y_1,\ldots,y_j,x_{j+1},\ldots,x_s|x'). \tag{12}$$

Bearing in mind the definition of the reduced density matrix, Eq. (2), we obtain

$$P^{(N)}_s(x|x') = A_x\sum_{j=0}^{k}(-1)^j\binom{s}{j}\int_y P^{(M)}_j(x_1,x_2,\ldots,x_j|y)Q^{(r)}_s(y,x_{j+1},\ldots,x_s|x'). \tag{13}$$

The integration over y is accomplished by substituting for $Q^{(r)}_s$ an expression of the form (10)

$$P^{(N)}_s(x|x') = \sum_{j=0}^{k}(-1)^j\binom{s}{j}A_xA_{x'}[P^{(M)}_j(x_1,x_2,\ldots,x_j|x_1',x_2',\ldots,x_j'),$$
$$p(x_{j+1}|x'_{j+1}),\ldots,p(x_s|x_s')]. \tag{14}$$

We state this final result as a separate theorem (assuming $s \leq M$).

THEOREM. Let $P^{(M)}_s(x|x')$ be a reduced density matrix representable by an antisymmetric wave-function of $M = r - N$ particles of rank r, u, and let $P^{(M)}_j(x|x') = \frac{j+1}{M-j}\int_y P^{(M)}_{j+1}(xy|x'y)$ be reduced density matrices of lower orders ($j < s$). The necessary and sufficient condition for the N-representability of the function $P_s^{(N)}(x|x')$ by a pure state of rank r is that it be expressible in the form of Eq. (14) through $P_s^{(M)}(x|x')$ and $P_s^{(M)}(x|x')$.

The necessary condition follows from the fact that if there exists a $\Psi_{(N)}(x)$ representing $P_s^{(N)}(x|x')$, then the $\Phi_{(M)}(y)$ related to it through Eq. (7) leads to reduced density matrices $P_j^{(M)}(x|x')$ satisfying Eq. (14). If there exists a $\Phi_{(M)}(y)$ representing $P_s^{(M)}(x|x')$ and, consequently $P_j^{(M)}(x|x')$ derived from it, then $\Psi_{(N)}(x)$, calculated according to Eq. (8), represents the $P_s^{(N)}(x|x')$ on the left side of Eq. (14).

2. The majority of known conditions for N-representability follow from Eq. (14).

For a fixed M and $r \to \infty$ the right side of Eq. (14) (as well as the left) is a kernel of a positive-definite operator. In particular, with s = 1 and s = 2 the right side of Eq. (14) contains kernels of the operators $g(x_1|x_1')$ and $Q(x_1, x_2|x_1', x_2')$ of [11], whose positive definiteness, it is maintained, is a "necessary" property on which several results of the above-mentioned paper are based. Thus, Eq. (14) contains a generalization of the Percus and Garrod inequalities for a reduced density matrix of higher order.

If we calculate the mean value of both sides of Eq. (14) for s = 2 with the determinant $\frac{1}{\sqrt{2}}\begin{vmatrix}\varphi_i(x_1)\varphi_i(x_2)\\ \varphi_j(x_1)\varphi_j(x_2)\end{vmatrix}$, the positiveness of the right side reduces to one of the necessary conditions for the N-representability formulated in [7].

Case of N-Representability of $P_1^{(N)}$ by a Pure State. We rewrite Eq. (14) for s = 1 in the form

$$P_1^{(N)}(x|x') + P_1^{(M)}(x|x') = p(x|x'). \tag{15}$$

If the rank r = N + 1, i.e., M = 1 and $P_1^{(M)}(x|x') = \varphi_1(x)\varphi_1^*(x')$, the Eq. (15) gives the natural $P_1^{(N)}(x|x')$ in the form

$$P_1^{(N)}(x|x') = \sum_{i=1}^{N+1} \varphi_i(x)\varphi_i^*(x') - \varphi_1(x)\varphi_1^*(x') = \sum_{i=2}^{N+1} \varphi_i(x)\varphi_i^*(x'). \tag{16}$$

But this expansion, consisting of N terms, is the first-order reduced density matrix for the determinant of N particles. Thus $\Psi_{(N)}(x)$ is a determinant, and one of the basis functions (φ_1) is not its natural one-particle state in contradiction to the original assumption, so that the rank of $\Psi_{(N)}(x)$ is N, and for a system of N-particles the rank N + 1 is not possible. This theorem was established by Coleman [1], and proved by Ando [2] and Foldy [10].

As was shown in [1], natural occupation numbers of first order for a two-particle system are even degenerate. Therefore, according to Eq. (15) and for an arbitrary even N and r = N + 2 the first-order natural occupation numbers are even degenerate. For N (and consequently for r = N + 2) odd in $P_1^{(N)}(x|x')$ there is one natural one-particle state which is not simultaneously the same for $P_1^{(2)}(x|x')$. Consequently, it belongs to unity. Such a structure of the spectrum of $P_1^{(N)}(x|x')$ for r = N + 2 was formulated by Coleman [1] as a necessary and sufficient condition for the N-representability of $P_1^{(N)}(x|x')$ by a pure state. The proof of the necessity which we obtained as a simple consequence of Eq. (14) was given by Ando [2] by a very tedious method, which required two preliminary lemmas.

On the other hand, let a first-order reduced density matrix be given with the above spectral structure

$$P_1^{(N)}(x|x') = \sum_{i=1}^{n+1} \Lambda_i[\varphi_i(x)\varphi_i^*(x') + \chi_i(x)\chi_i^*(x')] + \begin{cases} \varphi_0(x)\varphi_0^*(x') & \text{for N odd,} \\ 0 & \text{for N even,} \end{cases} \qquad n = \left[\frac{N}{2}\right]. \tag{17}$$

It is easy on the basis of the stated theorem to construct the wavefunction representing $P_1^{(N)}(x|x')$. For this we compute $P_1^{(2)}(x|x')$ from Eq. (15) and construct a two-particle function $\Phi_2(y)$ with first-order reduced density matrix $P_1^{(2)}(x|x')$, after which $\Psi_{(N)}(x)$ is determined from Eq. (8):

$$\Psi_{(N)}(x) = \sum_{i=1}^{n+1} \sqrt{1-\Lambda_i}\det[\varphi_1, \chi_1, \ldots, \varphi_{i-1}, \chi_{i-1}, \varphi_{i+1}, \chi_{i+1}, \ldots, \varphi_{n+1}, \chi_{n+1}](x) \tag{18}$$

(N even). For N odd $\Psi_{(N)}(x)$ has the same form of Eq. (18), but each determinant must include also φ_0. For superposition of functions of the type $\Psi_{(N)}(x)$ with different mutually orthogonal φ_0, using Eq. (17), it is easy to obtain Theorem 3.6 from [8].

Equation (18) not only verifies the sufficiency, but also shows incidentally that for a rank N + 2 the minimum number of determinants from which the wavefunction can be constructed is [N/2] + 1. Thus, Eq. (18) supports the solution found by Ando [2] for the problem posed by Foldy [10].

From Eq. (15) follows one more well-known specific case of the N-representability of $P_1^{(N)}(x|x')$. Let $P_1^{(N)}(x|x')$ have k natural occupation numbers equal to 1:

$$P_1^{(N)}(x|x') = \sum_{i=1}^{k} \varphi_i(x)\varphi_i^*(x') + P_1(x|x'). \tag{19}$$

Then the "remainder" $P_1(x|x')$ represents a first-order reduced density matrix represented by a function of N − k particles, strongly orthogonal to each of the one-electron functions $\varphi_1, \varphi_2, \ldots, \varphi_k$. To show that, we substitute Eq. (19) into Eq. (15):

$$P_1^{(M)}(x|x') + P_1(x|x') = p_r(x|x') - \sum_{i=1}^{k} \varphi_i(x)\varphi_i^*(x') = p_{r-k}(x|x'), \tag{20}$$

where p_{r-k} is a "unity" of the $(r-k)$-dimensional subspace orthogonal to $\varphi_1, \varphi_2, \ldots, \varphi_k$. It is evident that $\varphi_1, \varphi_2, \ldots, \varphi_k$ cannot be eigenfunctions of $P_1^{(M)}(x|x')$, since further application of Eq. (15) to $P_1^{(M)}(x|x')$ gives

$$P_1^{(M)}(x|x') + P_1^{(r-k-M)}(x|x') = p_{r-k}(x|x'), \tag{21}$$

where $P_1^{(r-k-M)}(x|x') - r - k - M = N$ is a k-representable reduced density matrix with natural states orthogonal to $\varphi_1, \varphi_2, \ldots, \varphi_k$. Comparing Eqs. (20) and (21), we have $P_1(x|x') = P_1^{(N-k)}(x|x')$, which proves the statement. (The theorem presented in [1] without proof contains an improper normalization $P_1^{(N-k)} \cdot (x|x')$.)

3. The main significance of Eq. (14) is in the fact that it allows to construct N-representable reduced density matrices for a system with a large number of particles when reduced density matrices of the same order but representable by functions of a small number of particles are available. For $s > 1$ the situation is substantially more complicated in comparison with the case $s = 1$, since due to the presence of lower reduced density matrices on the right side of Eq. (14) it is not diagonal in the basis of natural s-particle states of the function $\Phi_{(M)}(x)$. However, for specific values of the rank it is possible even here to determine a detailed construction of the reduced density matrix.

Let us, for example, find the natural expansion of the first-order reduced density matrix with rank $N + 2$. In this case $P_1^{(N)}(x|x')$ must have the form of Eq. (17) according to what has been proved. For simplicity we assume that there is no additional degeneracy among the Λ_i. Then for N even $P_2^{(N)}(x|x')$ has $n(n+1)/2$ natural occupation numbers $\Lambda_i + \Lambda_j - 1$ $(i \neq j)$, to each of which belong four linear combinations of determinants constructed from one-electron functions taken from two pairs $\varphi_i\chi_i$ and $\varphi_j\chi_j$, and $(n+1)$ natural occupation numbers ξ_k, which are solutions of the equation

$$1 + \frac{1-\Lambda_1}{2\Lambda_1 - 1 - \xi} + \frac{1-\Lambda_2}{2\Lambda_2 - 1 - \xi} + \ldots + \frac{1-\Lambda_{n+1}}{2\Lambda_{n+1} - 1 - \xi} = 0. \tag{22}$$

The natural two-particle states corresponding to ξ_k are linear combinations of the functions $\frac{1}{\sqrt{2}} \begin{vmatrix} \varphi_i(x_1)\varphi_i(x_2) \\ \chi_i(x_1)\chi_i(x_2) \end{vmatrix}$, $i = 1, 2, \ldots, n+1$. For N odd, there are in addition $n+1$ doubly degenerate natural occupation numbers $1 - \Lambda_i$ to each of which belong

$$\frac{1}{\sqrt{2}} \begin{vmatrix} \varphi_0(x_1)\,\varphi_0(x_2) \\ \varphi_i(x_1)\,\varphi_i(x_2) \end{vmatrix} \quad \text{and} \quad \frac{1}{\sqrt{2}} \begin{vmatrix} \varphi_0(x_1)\,\varphi_0(x_2) \\ \chi_i(x_1)\,\chi_i(x_2) \end{vmatrix}.$$

All this is the result of calculating the first-order reduced density matrix for the function (18) and of diagonalizing this expression.

It is interesting to note that the left side of Eq. (22) changes sign each time ξ goes through $2\Lambda_i - 1$, and therefore the roots are located in the intervals

$$2\Lambda_{n+1} - 1 < \xi_{n+1} < 2\Lambda_n - 1 < \xi_n < \ldots < \xi_2 < 2\Lambda_1 - 1 < \xi_1. \tag{23}$$

Since $2\Lambda_i - 1 \leq 1$, no natural occupation numbers of $P_2^{(N)}(x|x')$ with the possible exception of ξ_1 exceed unity. As was shown by us [12], this possibility of one of the occupation numbers increasing leads also for extremal functions of higher rank to $\xi_1 \sim N/2$, which implies a "nondiagonal long-range order" and superconductivity [13].

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