Separability of Very Noisy Mixed States and Implications for NMR Quantum Computing

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We give a constructive proof that all mixed states of N qubits in a sufficiently small neighborhood of the maximally mixed state are separable (unentangled). The construction provides an explicit representation of any such state as a mixture of product states. We give upper and lower bounds on the size of the neighborhood, which show that its extent decreases exponentially with the number of qubits. The bounds show that no entanglement appears in the physical states at any stage of present NMR experiments. Though this result raises questions about NMR quantum computation, further analysis would be necessary to assess the power of the general unitary transformations, which are indeed implemented in these experiments, in their action on separable states.

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In this Letter we investigate the structure of the space of density matrices of N spin-$1/2$ particles (qubits). In particular, we consider density matrices that are close to the maximally mixed density matrix and ask whether or not they are separable. A separable density matrix is one that can be written as a mixture of direct-product states. The statistics of all measurements made on a separable state of N qubits can be understood in terms of classical correlations among spin directions. Thus a separable state has no quantum entanglement. It has been argued that entanglement is the essential resource that gives a quantum computer its enhanced information-processing power [1].

One might imagine that the question addressed here is straightforward, in that the maximally mixed state seems to be very far from the boundary between separable and nonseparable states. It might be the case, however, that the maximally mixed density matrix is surrounded by separable matrices, but that these separable density matrices lie in a low-dimensional subspace within the space of all density matrices. By leaving this subspace, even infinitesimally, one could reach entangled density matrices. In Ref. [2] this problem is addressed by an existence proof; namely, it is shown that there exists a sufficiently small neighborhood of the maximally mixed density matrix inside which all density matrices are separable. In Ref. [3] a lower bound on the size of the neighborhood is given. Here we go further by giving a constructive proof that provides an explicit representation of any state sufficiently close to the maximally mixed one as a mixture of product states. We give an upper bound and a much improved lower bound on the size of the neighborhood, which show that the size decreases exponentially with the number of qubits.

Our results have immediate implications for present research that makes use of high-temperature, liquid-state nuclear magnetic resonance (NMR) for quantum-information processing and quantum computation [4–15]. Since the first proposals to use NMR for quantum computation, there has been surprise about the apparent ability to perform quantum computations in room-temperature thermal ensembles. It has been a puzzle how these thermal states, which are very close to the maximally mixed state, could correspond to truly entangled states [16]. The bounds we calculate show that *all states so far used in NMR for quantum computations or for other quantum-information protocols are separable.* This does not mean that NMR techniques are incapable of producing entangled states, in principle. Increasing the number of correlated spins might lead to nonseparable states, but this question is left open by the bounds derived in this paper.

We consider arbitrary density matrices for N qubits, written as

$$\rho_\epsilon = (1 - \epsilon)M_d + \epsilon\rho_1 , \tag{1}$$

where $d = 2^N$ is the Hilbert-space dimension for N qubits, $M_d = 1_d/d$ is the maximally mixed density matrix (1_d is the identity matrix in d dimensions), and ρ_1 is an arbitrary density matrix. Any density matrix can be written in the form (1). In the NMR context there are a macroscopic number of molecules in the liquid sample, each containing N active nuclear spins, and the density matrix (1) describes the state of each molecule. We show that for ϵ sufficiently small, all density matrices of the form (1) are separable. We define two kinds of representations of ρ_ϵ in terms of product states, which provide candidates for ensemble decompositions of ρ_ϵ as a mixture of product states. By considering these candidate decompositions, we derive an explicit lower bound on the size of the neighborhood of separable states. We conclude by establishing an explicit upper bound on the size of the neighborhood.

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Our approach is to represent an arbitrary density matrix in an overcomplete matrix basis, each basis element of which is a pure direct-product density matrix. If all the coefficients of a density matrix in this representation are non-negative, the coefficients can be considered to represent probabilities, and the density matrix is separable, as it is then a mixture of direct products.

All of our representations arise ultimately from expanding a density matrix for N qubits in terms of direct products of Pauli matrices:

$$\rho = \frac{1}{2^N} c_{\alpha_1 \ldots \alpha_N} \sigma_{\alpha_1} \otimes \cdots \otimes \sigma_{\alpha_N} . \tag{2}$$

Here and throughout we sum over repeated indices: Greek indices run over the values 0, 1, 2, 3, and Latin indices take on the values 1, 2, 3. The matrix $\sigma_0 = 1_2$ is the two-dimensional identity matrix, and the matrices σ_i, $i = 1, 2, 3$, are the Pauli matrices. The (real) expansion coefficients in Eq. (2) are given by the expectation values

$$\mathrm{tr}(\rho \sigma_{\alpha_1} \otimes \cdots \otimes \sigma_{\alpha_N}) = c_{\alpha_1 \ldots \alpha_N} . \tag{3}$$

Normalization requires that $c_{0\cdots 0} = 1$. Since the eigenvalues of the Pauli matrices are ± 1, the expansion coefficients satisfy $-1 \leq c_{\alpha_1 \cdots \alpha_N} \leq 1$.

To be concrete, we consider first the case of two qubits. For each qubit we introduce six pure density matrices, $P_i \equiv \frac{1}{2}(1_2 + \sigma_i)$ and $\overline{P}_i \equiv \frac{1}{2}(1_2 - \sigma_i)$. A convenient discrete overcomplete basis for discussing separability consists of the 36 direct-product projectors, each of which is a pure direct-product density matrix: $P_i \otimes P_j$, $P_i \otimes \overline{P}_j$, $\overline{P}_i \otimes P_j$, $\overline{P}_i \otimes \overline{P}_j$. Any density matrix of two qubits can be expanded in this basis, but since the basis is overcomplete, the representation is not unique. We make a specific choice, as follows. Noting that $\sigma_i = P_i - \overline{P}_i$ and $1_2 = P_i + \overline{P}_i$, we can write $1_2 = \omega_i(P_i + \overline{P}_i)$, where $\omega_i = 1/3$, $i = 1, 2, 3$. With these results we can convert the Pauli representation (2) into the form

$$\begin{aligned}\rho = \frac{1}{4}[&(\omega_i \omega_j + c_{i0}\omega_j + \omega_i c_{0j} + c_{ij}) P_i \otimes P_j + (\omega_i \omega_j - c_{i0}\omega_j + \omega_i c_{0j} - c_{ij}) \overline{P}_i \otimes P_j \\ &+ (\omega_i \omega_j + c_{i0}\omega_j - \omega_i c_{0j} - c_{ij}) P_i \otimes \overline{P}_j + (\omega_i \omega_j - c_{i0}\omega_j - \omega_i c_{0j} + c_{ij}) \overline{P}_i \otimes \overline{P}_j]. \end{aligned} \tag{4}$$

If the coefficient of each of the 36 basis elements is non-negative, the density matrix is separable. We note that when the maximally mixed density matrix for two qubits, $M_4 = \frac{1}{4} 1_2 \otimes 1_2$, is represented as in Eq. (4), the coefficient of each of the basis matrices is $1/36$.

Consider now an arbitrary entangled (nonseparable) density matrix ρ_1. Since ρ_1 is entangled, at least one of the coefficients in the representation of ρ_1 in the form (4) is negative. Suppose now that ρ_1 is mixed with the maximally mixed density matrix M_4 as in Eq. (1), i.e., $\rho_\epsilon = (1 - \epsilon) M_4 + \epsilon \rho_1$. Although some of the coefficients of ρ_1 are negative, all of the coefficients of M_4 are strictly positive. Hence, for ϵ small enough, all the coefficients of ρ_ϵ are non-negative, making ρ_ϵ separable. Thus *all* density matrices in a sufficiently small neighborhood of the maximally mixed density matrix are separable.

Furthermore, we can find an explicit bound on ϵ such that ρ_ϵ is separable for any ρ_1. To find a bound, we use $|c_{\alpha_1 \alpha_2}| \leq 1$ to bound the coefficients in a representation of ρ_1 of the form (4). The minimum value of any of the coefficients is $(1/4)(1/9 - 1/3 - 1/3 - 1) = -14/36$. Thus all the coefficients of the density matrix ρ_ϵ in the discrete overcomplete basis are non-negative if $(1 - \epsilon)/36 - 14\epsilon/36 \geq 0$, i.e., if $\epsilon \leq 1/15$. For $\epsilon \leq 1/15$, the representation (4) is an explicit decomposition of ρ_ϵ as a mixture of direct products.

A similar analysis can be carried out for any number of qubits. Starting from the Pauli representation (2), we introduce a discrete product basis, like that for two qubits, and define a representation analogous to that in Eq. (4). Using $|c_{\alpha_1 \ldots \alpha_N}| \leq 1$ to limit the size of the coefficients in this representation, we find an asymptotic lower bound on the size of the neighborhood of separable density matrices that is of order $\epsilon \sim 1/4^N$ for N qubits.

We turn now to another overcomplete basis for the space of density matrices, a basis labeled by continuous parameters. An arbitrary density matrix for N qubits can be represented as

$$\rho = \int d\Omega_1 \cdots d\Omega_N \, w(\vec{n}_1, \ldots, \vec{n}_N) P_{\vec{n}_1} \otimes \cdots \otimes P_{\vec{n}_N} , \tag{5}$$

where the integral runs over N Bloch spheres and where $P_{\vec{n}} \equiv \frac{1}{2}(1_2 + \vec{n} \cdot \vec{\sigma})$ is the projector onto the pure state located at unit vector $\vec{n}$. The representation (5) is by no means unique. In a spherical-harmonic expansion of $w(\vec{n}_1, \ldots, \vec{n}_N)$, the density matrix determines only the $l = 0$ and $l = 1$ components; the higher-order spherical harmonics correspond to the freedom in representing ρ as a sum of one-dimensional product projectors. A separable density matrix is one for which there exists a non-negative $w(\vec{n}_1, \ldots, \vec{n}_N)$, which can thus be interpreted as a probability density. In terms of the representation (5), the expectation values (3) are given by

$$\begin{aligned} c_{\alpha_1 \ldots \alpha_N} = \int & d\Omega_1 \cdots d\Omega_N \, w(\vec{n}_1, \ldots, \vec{n}_N) \\ & \times (n_1)_{\alpha_1} \cdots (n_N)_{\alpha_N} , \end{aligned} \tag{6}$$

where $(n_j)_0 = 1$. This illustrates that if the state is separable, the statistics of measurements can be understood in terms of classical correlations among spin directions.

We can generate a candidate for a separable ensemble decomposition of ρ by considering the unique representation of the form (5) such that $w(\vec{n}_1, \ldots, \vec{n}_N)$ has only $l = 0$ and $l = 1$ components. We can obtain this unique representation by noting that

$$\frac{1}{2}\,\sigma_\alpha = \frac{3}{4\pi}\int d\Omega\,\bar{n}_\alpha P_{\vec{n}}\,, \tag{7}$$

where $\bar{n}_0 = 1/3$ and $\bar{n}_j = n_j$. Inserting this result into the Pauli-matrix expansion (2) and using Eq. (3) gives

$$\begin{aligned} w(\vec{n}_1,\ldots,\vec{n}_N) &= \left(\frac{3}{4\pi}\right)^N c_{\alpha_1\ldots\alpha_N}(\bar{n}_1)_{\alpha_1}\cdots(\bar{n}_N)_{\alpha_N} \\ &= \frac{1}{(4\pi)^N}\,\mathrm{tr}[\rho(1_2 + 3\vec{n}_1\cdot\vec{\sigma}) \\ &\quad \otimes\cdots\otimes(1_2 + 3\vec{n}_N\cdot\vec{\sigma})]\,. \end{aligned} \tag{8}$$

The maximally mixed density matrix, M_{2^N}, has $w = (1/4\pi)^N$. The representation (8) has been considered previously by Scully and Wódkiewicz [17].

Let us concentrate on the operator product in the last form of Eq. (8). Each operator in the product has eigenvalues 4 and -2. Thus the most negative eigenvalue of the operator product is $4^{N-1}(-2) = -2^{2N-1}$, which implies that

$$w(\vec{n}_1,\ldots,\vec{n}_N) \ge -\frac{2^{2N-1}}{(4\pi)^N}\,. \tag{9}$$

Consider now the density matrix (1). Its candidate ensemble probability satisfies

$$\begin{aligned} w_\epsilon(\vec{n}_1,\ldots\vec{n}_N) &= \frac{1-\epsilon}{(4\pi)^N} + \epsilon w_1(\vec{n}_1,\ldots,\vec{n}_N) \\ &\ge \frac{1-\epsilon(1+2^{2N-1})}{(4\pi)^N}\,. \end{aligned} \tag{10}$$

Therefore ρ_ϵ is separable if

$$\epsilon \le \frac{1}{1+2^{2N-1}} \underset{N\to\infty}{\sim} \frac{2}{4^N}\,. \tag{11}$$

We see again that all density matrices in the neighborhood of the maximally mixed density matrix are separable, and we obtain a lower bound on the size of the separable neighborhood. For $N \ge 4$ our bound is better than the bound $\epsilon \le 1/(1+2^{N-1})^{(N-1)}$, given in Ref. [3].

One particularly interesting example is the Greenberger-Horne-Zeilinger (GHZ) state [13,18], a state of three qubits with density matrix

$$\begin{aligned} \rho_{\mathrm{GHZ}} = \frac{1}{2}(|111\rangle + |222\rangle)(\langle 111| + \langle 222|) = \frac{1}{8}(&1_2\otimes 1_2\otimes 1_2 + 1_2\otimes\sigma_3\otimes\sigma_3 + \sigma_3\otimes 1_2\otimes\sigma_3 \\ &+ \sigma_3\otimes\sigma_3\otimes 1_2 + \sigma_1\otimes\sigma_1\otimes\sigma_1 - \sigma_1\otimes\sigma_2\otimes\sigma_2 \\ &- \sigma_2\otimes\sigma_1\otimes\sigma_2 - \sigma_2\otimes\sigma_2\otimes\sigma_1)\,, \end{aligned} \tag{12}$$

for which Eq. (8) gives a representation

$$w_{\mathrm{GHZ}}(\vec{n}_1,\vec{n}_2,\vec{n}_3) = \frac{1}{(4\pi)^3}[1 + 9(c_1c_2 + c_2c_3 + c_1c_3) + 27s_1s_2s_3\cos(\varphi_1+\varphi_2+\varphi_3)] \ge -\frac{26}{(4\pi)^3}\,. \tag{13}$$

Here $c_j \equiv \cos\theta_j$ and $s_j \equiv \sin\theta_j$, and the minimum occurs at $\theta_1 = \theta_2 = \theta_3 = \pi/2$ and $\varphi_1 + \varphi_2 + \varphi_3 = \pi$. Thus the mixed state $\rho_\epsilon = (1-\epsilon)M_8 + \epsilon\rho_{\mathrm{GHZ}}$ is separable if $\epsilon \le 1/27$, in which case no measurement can reveal evidence of quantum entanglement.

Up to this point we have been thinking of the number of qubits as being fixed, and we have investigated the boundary between separability and nonseparability as the amount of noise, specified by ϵ, changes. We now shift gears, thinking of the qubits as particles with spin and asking what happens as the number of particles or their dimension changes, while ϵ is held fixed. In general, as we go to more particles or higher spins, we find that we can tolerate more mixing with the maximally mixed state and still have states that are not separable. In other words, for a given ϵ, we can always find states of sufficiently large numbers of particles or sufficiently high spin for which ρ_ϵ is nonseparable. We translate this result into an upper bound on the size of the separable neighborhood around the maximally mixed state.

Consider now two spin$-(d-1)/2$ particles, each living in a d-dimensional Hilbert space. What we have in mind is that each of these particles is an aggregate of $N/2$ spin-$1/2$ particles (qubits), in which case $d = 2^{N/2}$. We consider a specific joint density matrix of the two particles,

$$\rho_\epsilon = (1-\epsilon)M_{d^2} + \epsilon|\psi\rangle\langle\psi|\,, \tag{14}$$

where $|\psi\rangle$ is a maximally entangled state of the two particles,

$$|\psi\rangle = \frac{1}{\sqrt{d}}(|1\rangle|1\rangle + |2\rangle|2\rangle + \cdots + |d\rangle|d\rangle)\,. \tag{15}$$

Now project each particle onto the subspace spanned by $|1\rangle$ and $|2\rangle$. The state after projection is

$$\tilde{\rho} = \frac{1}{A}\left(\frac{1-\epsilon}{d^2}1_4 + \frac{\epsilon}{d}(|1\rangle|1\rangle + |2\rangle|2\rangle)(\langle 1|\langle 1| + \langle 2|\langle 2|)\right) = (1-\epsilon')M_4 + \epsilon'|\phi\rangle\langle\phi|\,, \tag{16}$$

where $A = (4/d^2)[1 + \epsilon(d/2-1)]$ is the normalization factor,

$$|\phi\rangle = \frac{1}{\sqrt{2}}(|1\rangle|1\rangle + |2\rangle|2\rangle) \tag{17}$$

is a maximally entangled state of two qubits, and

$$\epsilon' = \frac{2\epsilon/d}{A} = \frac{\epsilon d/2}{1 + \epsilon(d/2 - 1)}. \tag{18}$$

The projected state $\tilde{\rho}$ is a Werner state [19], a mixture of the maximally mixed state for two qubits, M_4, and the maximally entangled state $|\phi\rangle$. The proportion ϵ' of maximally entangled state increases linearly with d. Thus, as d increases for fixed ϵ, there is a critical dimension beyond which $\tilde{\rho}$ becomes entangled. Indeed, the Werner state is nonseparable for $\epsilon' > 1/3$ [19,20], which is equivalent to $d > \epsilon^{-1} - 1$. Moreover, since the local projections on the two particles cannot create entanglement from a separable state, we can conclude that the state (14) of N qubits is nonseparable under the same conditions, i.e., if [21]

$$\epsilon > \frac{1}{1 + d} = \frac{1}{1 + 2^{N/2}}. \tag{19}$$

This result establishes an *upper* bound, scaling like $2^{-N/2}$, on the size of the separable neighborhood around the maximally mixed state.

Our results have implications for attempts to use high-temperature NMR techniques to perform quantum computations or other quantum-information-processing tasks. They imply that NMR experiments performed to date have not produced genuinely entangled density matrices. This is because in current experiments, the parameter ϵ, which measures the deviation from the maximally mixed state, has a value $\sim 3 \times 10^{-5}$, much smaller than the lower bounds we have found for the radius of the separable neighborhood of the maximally mixed state, for the cases of two or three spins used in these experiments.

Present high-temperature NMR techniques, based on synthesizing a pseudopure state, give an ϵ that scales like $N/2^N$ as the number of qubits increases at constant temperature [4,6]. With this scaling, the state ρ_ϵ leaves the region where our lower bound implies that all states are separable at about 13 qubits, but it never enters the region where our upper bound guarantees that there are entangled states. Thus, it is unclear whether present NMR techniques can produce entangled states. Different techniques might lead to a more favorable scaling behavior for ϵ [22].

The results in this Letter suggest that current NMR experiments are not true quantum computations, since no entanglement appears in the physical states at any stage [1]. We stress, however, that we have not proved this suggestion, since we would need to analyze the power of general unitary operations, which are successfully implemented in these experiments, in their action on separable states [23]. To reach a firm conclusion, much more needs to be understood about what it means for a computation to be a "quantum" computation.

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