

Novel Strategy for Database Searching in Spin Liouville Space by NMR Ensemble Computing

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Quantum computing by nuclear magnetic resonance using pseudopure spin states is bound by the maximal speed of quantum computing algorithms operating on pure states. In contrast to these quantum computing algorithms, a novel algorithm for searching an unsorted database is presented here that operates on truly mixed states in spin Liouville space. It provides an exponential speedup over Grover's quantum search algorithm with the sensitivity scaling exponentially with the number of spins, as for pseudopure state implementations. The minimal decoherence time required is exponentially shorter than that for Grover's algorithm.

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Nuclear magnetic resonance (NMR) with its highly developed radio-frequency pulse methods for generating a large variety of unitary evolutions [1] is currently the leading technology for the experimental realization of quantum computations (for a recent review, see [2]). Nearly all NMR quantum computations reported to date use a *pseudopure state* [3,4] as initial state represented by the density operator $\sigma = (1 - \varepsilon)2^{-n}\mathbf{1} + \varepsilon|00\cdots 0\rangle\langle 00\cdots 0|$, where n is the number of spin $\frac{1}{2}$ nuclei (= quantum bits or qubits) of a single molecule of the ensemble, $|00\cdots 0\rangle$ is usually the spin-ground state wave function of a single molecule, and $\mathbf{1}$ is the unity operator. Since the prefactor ε , which determines the sensitivity, decreases exponentially with the number of qubits, this approach is practical only for a restricted number of qubits [5,6]. Moreover, the maximal efficiency (computational speed) of this approach is the same as the one of a pure-state quantum computer. The actual "quantumness" of pseudopure states has recently been scrutinized [7,8].

Here, the potential of liquid-state NMR is expanded for performing certain computational tasks by using truly mixed spin states. Massive parallelism can potentially be achieved by representing input states by molecular subensembles possessing different spin states. In this way, molecules with different spin states simultaneously perform different computations. Like DNA computing [9], this kind of molecular parallelism is classical in nature, but unlike DNA computing, various linear combinations of different input states are prepared and evaluated.

Liouville space computation.—Recently, a formalism for NMR computations was introduced using density operators that are direct (tensor) products of spin-polarization operators to represent logic states [10]. Linear combinations of these states are generally truly mixed; i.e., they cannot be represented in Hilbert space but rather in spin Liouville space, which is the space that spans all conceivable density operators σ characterizing the spin ensemble. Unlike computations using pseudopure states, this scheme is not bound to the speedup limits of pure-state quantum computers as is demonstrated here

with a novel database search algorithm whose number of function evaluations scales with the logarithm of the number of database entries.

The 2^n eigenstates $|\psi\rangle$ of the Zeeman Hamiltonian created by a strong external magnetic field, which all have the form $|\psi\rangle = |001\cdots 01\rangle = |\alpha\alpha\beta\cdots\alpha\beta\rangle$ (α denotes spin "up" and β denotes spin "down"), are mapped on states in spin Liouville space [10]

$$|\psi_{\text{in}}\rangle = |\alpha\alpha\beta\cdots\alpha\beta\rangle \Rightarrow \sigma_{\text{in}} = |\psi_{\text{in}}\rangle\langle\psi_{\text{in}}| \\ = I_1^\alpha I_2^\alpha I_3^\beta \cdots I_{n-1}^\alpha I_n^\beta, \quad (1)$$

where $I_1^\alpha I_2^\alpha I_3^\beta \cdots I_{n-1}^\alpha I_n^\beta$ is the direct product of *polarization operators* I_k^α, I_k^β defined as [1]

$$I_k^\alpha = |\alpha_k\rangle\langle\alpha_k| = \frac{1}{2}(\mathbf{1}_k + 2I_{kz}) = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}, \\ I_k^\beta = |\beta_k\rangle\langle\beta_k| = \frac{1}{2}(\mathbf{1}_k - 2I_{kz}) = \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix}, \quad (2)$$

where $2I_{kz}$ is the Pauli matrix σ_z and $\mathbf{1}_k$ is the unity operator of the subspace of spin I_k . For what follows it is useful to note that $I_k^\alpha + I_k^\beta = \mathbf{1}_k$ and $I_k^\alpha - I_k^\beta = 2I_{kz}$. As common in NMR, σ_{in} is represented by $\gg 10^{12}$ molecules.

Database searching.—Grover's database search algorithm [11] is one of the few quantum-computing algorithms known to date considered to become potentially useful. It provides a quadratic speedup over classical approaches in the search of a single object in a large unsorted database. A common formulation of the problem is as follows. For a given binary function $f: \{0, 1\}^n \rightarrow \{0, 1\}$, which is known to be zero for all inputs \mathbf{x} , $f(\mathbf{x}) = 0$, except for one, say $\mathbf{x} = \mathbf{z}$, where $f(\mathbf{z}) = 1$. The problem is to find \mathbf{z} . On a classical computer one has to evaluate the function f on average about $(2^n + 1)/2$ times to find \mathbf{z} . In contrast, Grover's algorithm finds \mathbf{z} in $O(2^{n/2})$ steps, a speedup that is optimal for pure and pseudopure states [12].

In order to perform function f reversibly [13], an extra bit is required, represented by spin I_0 , which is at the beginning of the computation always in the α state.

Hence, the Liouville space representation of the input of f has the general form $I_0^\alpha \sigma_{\text{in}}$. The action of f can be described as a permutation of all states spanned by the $n + 1$ bits, which corresponds to a unitary transformation U (*vide infra*). With the output stored on spin I_0 , the result can be expressed in terms of the expectation value of operator I_{0z} ,

$$f = F(I_0^\alpha \sigma_{\text{in}}) = \frac{1}{2} - \text{Tr}\{UI_0^\alpha \sigma_{\text{in}} U^\dagger I_{0z}\}. \quad (3)$$

F can also be evaluated for a sum of M density operators, $\sum_{j=1}^M I_0^\alpha \sigma_j$, representing a truly mixed spin state, where σ_j is of the form of σ_{in} of Eq. (1):

$$f = \sum_{j=1}^M F(I_0^\alpha \sigma_j) = F\left(\sum_{j=1}^M I_0^\alpha \sigma_j\right) + \frac{(M-1)}{2}, \quad (4)$$

where the linearity of both the trace operation and the unitary transformation U with respect to $I_0^\alpha \sigma_j$ was used. Equation (4) shows how f can be evaluated simultaneously when applied to a linear combination of states in spin Liouville space.

So far, all experimental demonstrations of database searching using NMR have been based on pseudopure states [14–16]. It is shown in the following for a reversible implementation of f in spin Liouville space that an exponential speedup is possible. To describe the algorithm, it is useful to number all possible input states of Eq. (1) from 1 to $N = 2^n$: $\sigma_1 = I_1^\alpha I_2^\alpha \cdots I_{n-1}^\alpha I_n^\alpha$; $\sigma_2 = I_1^\alpha I_2^\alpha \cdots I_{n-1}^\alpha I_n^\beta$; $\sigma_3 = I_1^\alpha I_2^\alpha \cdots I_{n-1}^\beta I_n^\alpha$; $\sigma_4 = I_1^\alpha I_2^\alpha \cdots I_{n-1}^\beta I_n^\beta$; \dots ; $\sigma_N = I_1^\beta I_2^\beta \cdots I_{n-1}^\beta I_n^\beta$. Here and in the following the presence of the ancilla bit I_0^α as additional factor is tacitly assumed. Using Eq. (4) f is then evaluated on linear combinations of σ_j by following a “divide and conquer” scheme. First, f is evaluated on a state that is the sum of the first half of all possible states 1 through $N/2$. If the result is 1, then \mathbf{z} must be one of these states. To further localize \mathbf{z} , f is evaluated for the sum of states 1 through $N/4$. If the result is 1, then the procedure is repeated for the sum of states 1 through $N/8$. If, however, the result was 0, state \mathbf{z} must be one of the states $N/4 + 1$ to $N/2$ and the function is evaluated for the sum of states $N/4 + 1$ through $3N/8$. If the result is 0, then \mathbf{z} must be one of the states $3N/8 + 1$ to $N/2$. Next the function is evaluated for the sum of states $3N/8 + 1$ to $7N/16$, etc. At each step, one-half of the remaining candidates for \mathbf{z} are excluded. In this way, \mathbf{z} is tracked down in $n = \log_2 N$ steps. Thus, this algorithm offers an exponential speedup over a classical search and over Grover’s quantum algorithm.

The hierarchy of input states of the algorithm is depicted in Fig. 1(a) for $n = 4$ by a binary tree with four levels, $m = 0, 1, 2, 3$. The computation starts at the top of the figure (root $m = 0$) with I_1^α , which is the sum of the first eight states, $I_1^\alpha = \sum_{\alpha/\beta} I_1^\alpha I_2^{\alpha/\beta} I_3^{\alpha/\beta} I_4^{\alpha/\beta}$. At each step the result of the function evaluation determines which mixed state has to be generated as input for the following function evaluation: If $f = 1$ one proceeds to the left and if $f = 0$

one proceeds to the right. The states reached at the bottom give the results of the possible scenarios. The desired state \mathbf{z} , which is one out of $N = 2^4$ possible states, can be identified with exactly four function evaluations, while a classical computer requires on average 8.4375 function evaluations. The result of the function evaluation is determined by the sign of the (offset corrected) resonance of the readout spin I_0 : If it is -1 , then $f = 0$ (I_0^α); if it is $+1$, then $f = 1$ (I_0^β). If spin I_0 belongs to a different nuclear species than the other spins, Fourier transformation can be avoided by on-resonance recording of the integral of the free induction decay (FID). The evaluation of the result and the control of the pulse sequence for the preparation of the linear combination of states can be performed by a (standard) computer using, for example, a lookup table that contains the information of the binary tree of Fig. 1(a).

Although the same lookup table can be used for different database functions f , its size grows exponentially with n . Moreover, the farther down one climbs the binary tree of Fig. 1(a), the more sophisticated are the excitation schemes required to prepare the mixed input states. Fortunately, considerable simplification is possible by using a modified scheme [Fig. 1(b)]. It takes advantage of Eq. (4) by superimposing at each level m of the binary tree $2^{n-1} - 2^{(n-m-1)}$ additional suitably selected states to the input state. To illustrate the modified algorithm, let the computation again start with I_1^α with output $f = 1$. As above, it follows that \mathbf{z} is one of the eight states $I_1^\alpha I_2^{\alpha/\beta} I_3^{\alpha/\beta} I_4^{\alpha/\beta}$ and none of the remaining eight states $I_1^\beta I_2^{\alpha/\beta} I_3^{\alpha/\beta} I_4^{\alpha/\beta}$. Instead of using $I_1^\alpha I_2^\alpha$ as the next input, four of the eight states that were already ruled out, namely, $I_1^\beta I_2^\alpha I_3^\alpha I_4^\alpha$, $I_1^\beta I_2^\alpha I_3^\beta I_4^\beta$, $I_1^\beta I_2^\beta I_3^\alpha I_4^\alpha$, and $I_1^\beta I_2^\beta I_3^\beta I_4^\beta$, are added to $I_1^\alpha I_2^\alpha$. Thus, the next input state is $I_1^\alpha I_2^\alpha + I_1^\beta I_2^\alpha = I_2^\alpha$, which can be prepared analogously to I_1^α . Next, if the result is 0, f is applied to $I_1^\alpha I_2^\beta I_3^\alpha$ supplemented by the sum of the six states $I_1^\alpha I_2^\alpha I_3^\alpha I_4^{\alpha/\beta}$, $I_1^\beta I_2^\alpha I_3^\alpha I_4^{\alpha/\beta}$, $I_1^\beta I_2^\beta I_3^\alpha I_4^{\alpha/\beta}$, which yields I_3^α as the next input state. By repeating this argument it follows that the scheme of Fig. 1(a) can be replaced by the scheme of Fig. 1(b) only requiring function evaluations of input states $I_0^\alpha I_k^\alpha$, $k = 1, \dots, 4$. At all tree levels, the mixed input state contains at least $2^{n-1} - 1$ states for which $f = 0$. Since each of them yields the same negative offset to the resonance of spin I_0 , their net effect can be undone by adding a signal of relative magnitude $b = 2^{n-1} - 1$.

An important difference to the algorithm of Fig. 1(a) is the fact that on a given tree level all input states are the same and hence no logic decision has to be made concerning the preparation of the following input. A sequence of n experiments with inputs $I_0^\alpha I_k^\alpha$, $k = 1, \dots, n$, can be recorded, processed, and translated into a parity string of length n reflecting the amplitudes of the resonance of the detection spin I_0 in the n experiments: $+1$ signifies that \mathbf{z} contains I_k^α , while -1 signifies that \mathbf{z} contains I_k^β . Thus, the bits of \mathbf{z} can be sequentially identified by separate

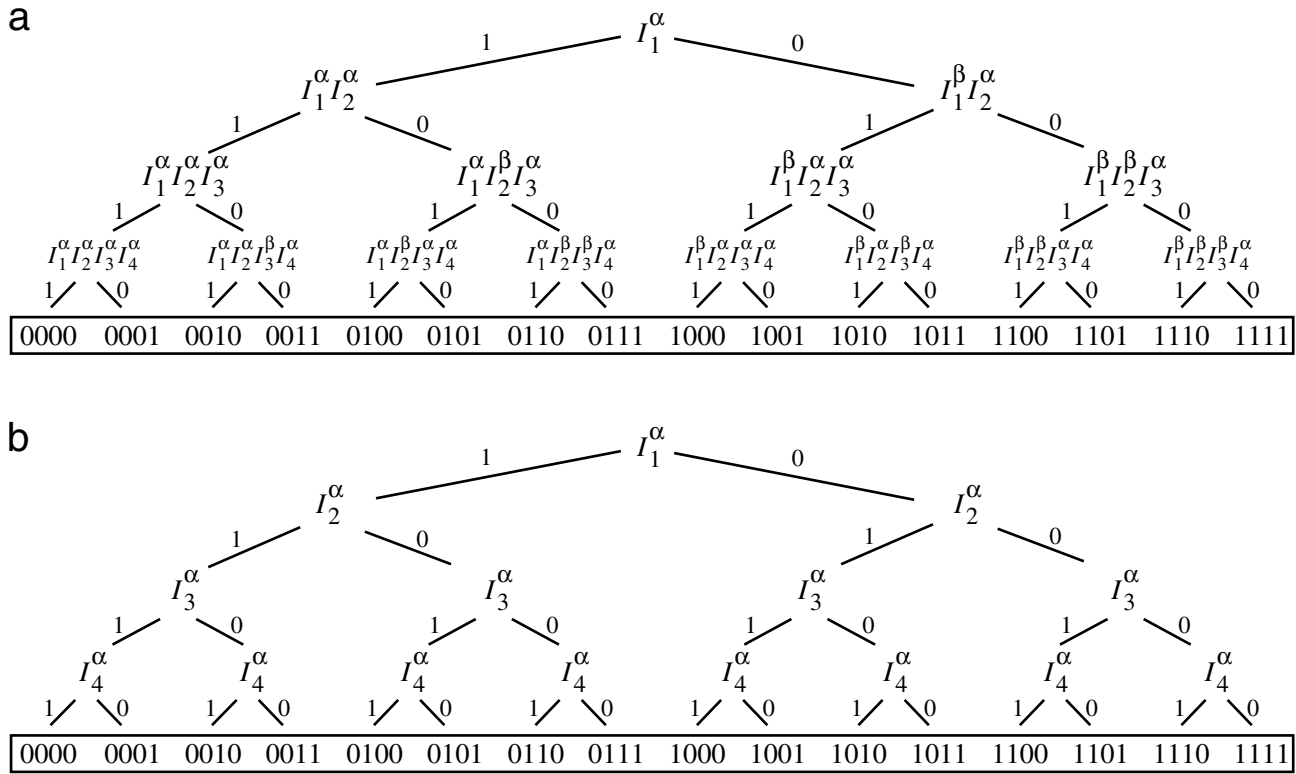


FIG. 1. Binary search trees for unsorted database search using truly mixed spin states in spin Liouville space. The nodes indicate the input states for the binary database search oracle function f . The computation starts at the top. All input states additionally include operator I_0^α as the factor representing an ancilla bit. (a) If $f = 1$ one proceeds down the tree to the next input state on the left-hand side, if $f = 0$ one proceeds to the right-hand side, and so on. The state reached at the bottom is the wanted database entry for which $f = 1$. (b) Search tree of an improved version of the algorithm of (a). Function evaluations on states of the type $I_0^\alpha I_k^\alpha$, which corresponds to suitable linear combinations of half of all possible input states and which are experimentally significantly easier to prepare than the input states of (a), lead to the same exponential speedup as the algorithm in (a).

experiments. The input states $I_0^\alpha I_k^\alpha$ (plus a multiple of $\mathbf{1}$, which will have no net effect on the result) can be readily prepared experimentally including the use of SWAP operations that interchange the states of spin pairs [10].

For a *sorted* database it is well known that a classical bisection method or a binary search algorithm provides an exponential speedup over a sequential or random search. In contrast, the Liouville space algorithm presented here yields an exponential speedup for the *unsorted* database. The parallelism is achieved by the evaluation of selected linear combinations of inputs in spin Liouville space, which fundamentally differs from classical parallelism as realized, for example, on a workstation cluster where the speedup for searching an unsorted database scales linearly with the number of workstations. Except for the ancilla prefactor I_0^α , the input states $I_0^\alpha I_k^\alpha$ correspond to the initial states of most multidimensional NMR experiments [1]. The role of these mixed states in the present context sheds additional light on the intrinsic power of NMR spectroscopy.

Example.—The search algorithm is illustrated here for a database with eight entries {000,001,010,011,100,101,110,111} represented by Zeeman eigenstates of the three spins I_1, I_2, I_3 . We define the database

function as $f = 1$ for entry $\mathbf{z} = 101$, represented by state $I_1^\beta I_2^\alpha I_3^\beta$, and $f = 0$ for the seven remaining entries {000,001,010,011,100,110,111} represented by the spin density operators $\{I_1^\alpha I_2^\alpha I_3^\alpha, I_1^\alpha I_2^\alpha I_3^\beta, I_1^\alpha I_2^\beta I_3^\alpha, I_1^\alpha I_2^\beta I_3^\beta, I_1^\beta I_2^\alpha I_3^\alpha, I_1^\beta I_2^\alpha I_3^\beta, I_1^\beta I_2^\beta I_3^\alpha, I_1^\beta I_2^\beta I_3^\beta\}$. With the use of the ancilla spin I_0 , function f can be realized by the permutation shown in Fig. 2. While we are not concerned here with the details of the experimental realization of the oracle f , it is noted that permutations of the kind of Fig. 2 can be achieved by a sequence of logic gates, which are realized using NMR by the combined use of radio-frequency pulses and J -coupling evolution [2,10,14–17]. Following the search scheme of Fig. 1(b), the first input state is $I_0^\alpha I_1^\alpha$, which is equal to the linear combination $I_0^\alpha I_1^\alpha I_2^\alpha I_3^\alpha + I_0^\alpha I_1^\alpha I_2^\alpha I_3^\beta + I_0^\alpha I_1^\alpha I_2^\beta I_3^\alpha + I_0^\alpha I_1^\alpha I_2^\beta I_3^\beta$. It will remain unchanged by the permutation of Fig. 2 and detection on spin I_0 will thus lead to a signal of relative strength -4 . Addition of $b = 2^{n-1} - 1 = 3$ yields a relative intensity -1 , which implies that the first bit of the wanted database entry must be 1 (I_1^β). To evaluate the second bit, f is applied to the input state $I_0^\alpha I_2^\alpha = I_0^\alpha I_1^\alpha I_2^\alpha I_3^\alpha + I_0^\alpha I_1^\alpha I_2^\alpha I_3^\beta + I_0^\alpha I_1^\beta I_2^\alpha I_3^\alpha + I_0^\alpha I_1^\beta I_2^\alpha I_3^\beta$, which according to Fig. 2 yields as output $I_0^\alpha I_1^\alpha I_2^\alpha I_3^\alpha + I_0^\alpha I_1^\alpha I_2^\alpha I_3^\beta + I_0^\alpha I_1^\beta I_2^\alpha I_3^\alpha +$

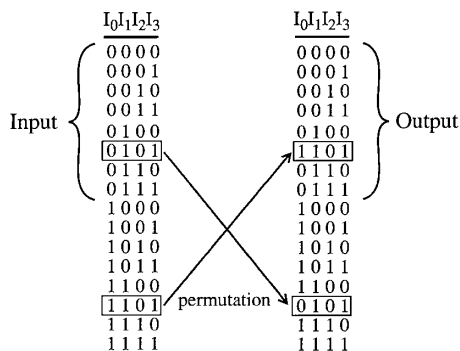


FIG. 2. Graphical representation of a particular database function f operating on spins I_1, I_2, I_3 as a permutation using an ancilla bit I_0 with the output stored on I_0 .

$I_0^\beta I_1^\beta I_2^\alpha I_3^\beta$. The first three terms yield -1 as output on spin I_0 , the fourth term yields $+1$, and hence the total intensity is -2 . The addition of $b = 3$ yields $+1$, which implies that the second bit must be 0 (I_2^α). Finally, the third bit is evaluated by applying f to $I_0^\alpha I_3^\alpha = I_0^\alpha I_1^\alpha I_2^\alpha I_3^\alpha + I_0^\alpha I_1^\alpha I_2^\beta I_3^\alpha + I_0^\alpha I_1^\beta I_2^\alpha I_3^\alpha + I_0^\alpha I_1^\beta I_2^\beta I_3^\alpha$ yielding again a total intensity of -4 on spin I_0 implying that the third bit is 1 (I_3^β). The three experiments together unambiguously identify the desired state $\mathbf{z} = 101$.

Sensitivity and decoherence time.—While the sensitivity of the input states of Fig. 1(b) does not depend on n , the readout of the result does. The fact that only one out of 2^n input states of Eq. (1) yields $f = 1$ implies that in the molecular ensemble on average one out of 2^n molecules exhibits this spin configuration and only the subensemble constituted of these molecules will eventually contribute to the readout signal. Thus, the sensitivity of the database search algorithm will decrease with 2^{-n} , which is the same as for implementations of Grover's algorithm using pseudopure states [14–16]. From the exponential speedup of the algorithm of Fig. 1 follows that a mixed-state implementation is clearly preferable over implementations using pseudopure states.

The exponential speedup can be converted into sensitivity enhancement by repeating the same computation and by accumulating the FID's. Since the signal-to-noise ratio S/N scales with the square root of the number of experiments, 2^{2n} experiments are required to compensate for the sensitivity loss of 2^{-n} . Thus, if for increasing n no sensitivity loss is tolerated, the total computation time scales with $n2^{2n}$ as compared to $2^{n/2}$ for a pure-state (and $2^{5n/2}$ for a pseudopure state) implementation of Grover's algorithm. In reality, however, high-field NMR spectrometers have a $S/N \gg 1$, which allows one to tolerate a certain sensitivity loss: For a given S/N , a database search in spin Liouville space is of advantage over Grover's search

with a single pure-state quantum computer for a maximal number qubits n fulfilling $\sqrt{n} 2^{3n/4} < S/N$. With current NMR spectrometer technology, this corresponds to two to three *quantum bytes*. For larger n a pure-state quantum computer would (if existent) currently be favorable.

A crucial factor for implementations of quantum algorithms is the minimal length required for the decoherence time (relaxation time) T_{\min} . For Grover's algorithm T_{\min} scales exponentially, $T_{\min} > 2^{n/2}t$, where t is the duration of a single evaluation of f . On the other hand, for the presented database search algorithm T_{\min} must exceed only the time required for a single evaluation of f , $T_{\min} > t$, which is a much less stringent criterion than the one for Grover's algorithm. This feature together with the exponential speedup makes spin Liouville space computation an attractive resource for ensemble computing in general.

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