Solving the quantum search problem in polynomial time on an NMR quantum computer

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Abstract

The quantum search problem is an important problem due to the fact that a general NP problem can be solved by an unsorted quantum search algorithm. Here it has been shown that the quantum search problem could be solved in polynomial time on an NMR quantum computer. The NMR ensemble quantum computation is based on the quantum mechanical unitary dynamics that both a closed quantum system and its ensemble obey the same quantum mechanical unitary dynamics instead of on the pseudopure state or the effective pure state of the classical NMR quantum computation. Based on the new principle the conventional NMR multiple-quantum spectroscopy has been developed to solve the search problem. The solution information of the search problem is first loaded on the unitary evolution propagator which is constructed with the oracle unitary operation and oracle-independent unitary operations and used to excite the multiple-quantum coherence in a spin ensemble. Then the multiple-quantum spectroscopy is used to extract experimentally the solution information. It has been discussed how to enhance the output NMR signal of the quantum search NMR multiple-quantum pulse sequence and some approaches to enhancing the NMR signal are also proposed. The present work could be helpful for the conventional high magnetic field NMR machines to solve efficiently the quantum search problem.

1. Introduction
An efficient quantum search algorithm may be tremendously useful because of the wide range of its practical applications, and especially due to the fact that a general NP problem can be solved by the unsorted quantum search algorithm [1]. It is believed extensively that a general NP problem can not be solved efficiently on a classical digital computer in polynomial time. But if there would be a polynomial-time quantum search algorithm, then a general NP problem would be efficiently solved by the algorithm in polynomial time on a quantum computer. In the past years a great effort has been devoted to searching for an efficient quantum search algorithm. On the basis of the quantum parallelism principle [2] Grover first proposed a quantum search algorithm with quadratic speedup over the classical counterparts [3, 4]. It has been shown that this algorithm is optimal [5, 6]. A large number of works of the Grover algorithm and its applications have been published in the past several years. However, the quantum search algorithms [3, 4] which are based on the pure quantum states are not polynomial-time algorithms and hence have not been proved certainly that they can move the frontier between solvability and intractability of complexity problems [6]. On the other hand, the experimental realization of quantum computation and the physical architecture of quantum computers have achieved a great progress in the recent years, although it is the long term to build up a commercial quantum computer. In particular, the experimental ensemble quantum computation based on the nuclear magnetic resonance (NMR) techniques [7, 8] has been extensively studied [9, 10] and a large number of the related works have been reported [11-14]. But the classical NMR quantum computation [9-15] working on the concept of the pseudopure state [9] or the effective pure state [10] suffers from many trouble problems [15, 16]. For example, people suspect whether or not the classical NMR quantum computation is real quantum computation and is as powerful as the quantum computation based on pure quantum states [16]. Since the effective pure states or the pseudopure states are isomorphic to the pure quantum states [9, 10, 12] the classical NMR quantum computation is at most as powerful as the original one based on pure quantum states and thus, can not be able to move the frontier between solvability and intractability. It has to give up many important characteristic features of a spin ensemble in order that with the help of the concept of the effective pure state or the pseudopure state the pure-state the pure-state quantum computation can work in the spin ensemble as well. For example, highly mixed states of a spin ensemble are not suitable for the classical NMR quantum computation. It is known that the quantum states
of the spin system in a spin ensemble are very highly mixed. Therefore, the
classical NMR quantum computation can not give a satisfied answer whether
or not the highly mixed states in a spin ensemble could be used in quantum
computation and about the power question of quantum computation with
the highly mixed states.

Amazingly, recently it has been shown that an NMR quantum computer
operating on the highly mixed states of a spin quantum ensemble can be
exploited to solve efficiently the unsorted search problem [17] and the parity
problem [18] in polynomial time, while these problems have been shown to
be hard NP problems and can not be solved efficiently in classical compu-
tation and even in pure-state quantum computation [19, 20]. These results
show powerfully that (a) a general NP problem could be solved in polyno-
mial time on a quantum computer; (b) quantum entanglement of a quantum
system may not be the sole origin of quantum computation power and the
quantum mechanical unitary dynamics may play an important role in solving
these NP problems in polynomial time; (c) quantum computers can outper-
form classical computers and ensemble quantum computation can be at least
as powerful as the standard quantum computation based on pure quantum
states. These quantum algorithms or sequences that operate on highly mixed
states in a spin quantum ensemble could achieve an exponential speedup over
the classical search computation and therefore they really move the frontier
between the solvability and intractability of complex problems. Although
in theory the NMR ensemble quantum computation on highly mixed states
is successful to solve the unsorted quantum search problem and the parity
problem in polynomial time, it still suffers from some problems and limits in
practice. The most severe problem is that the output NMR signal intensity of
these sequences reduces exponentially as the qubit number of the spin system
in the spin ensemble [17, 18], making the NMR ensemble quantum compu-
tation available only in spin systems with a small number of qubits. There
may be two schemes to extend the scale of the NMR quantum computation
so that it could solve the search problem with a larger qubit number. One
is to enhance the initial spin polarization of a spin ensemble by a variety of
possible spin-polarization enhancement techniques [7, 8, 21, 22, 23]. Another
is to design new quantum search algorithms or sequences which consist of a
polynomial number of the oracle unitary operations and oracle-independent
unitary operations since the output NMR signal could be increased by many
calls of the oracle unitary operations in the sequences. In this paper ac-
cording to the second scheme new quantum search sequences on the NMR
ensemble quantum computers are proposed to solve the search problem with the help of the multiple-quantum operator algebra space formalism [24, 25], and particularly the NMR multiple-quantum spectroscopy [7, 8] has been developed to search for the unknown marked state, i.e., the solution of the search problem.

There may be two possible schemes for quantum computation making a transition from a pure-state quantum system to its quantum ensemble. One is the classical NMR quantum computation. It is based on the concept of the effective pure state [10] or the pseudopure state [9] which is isomorphic to the pure quantum state. Then, the quantum computation on the pure state quantum systems allows to apply to their quantum ensembles with the help of the concept of the effective pure state. In the frame of the classical NMR quantum computation information of quantum computation is mainly carried by the effective pure states. Another is the new NMR ensemble quantum computation. It is based on the fact that both the closed pure-state quantum system and its quantum ensemble obey the same quantum mechanical unitary dynamics when decoherence effects in the quantum system and its ensemble are negligible [17, 18]. Then in the frame of the NMR ensemble quantum computation the information of quantum computation is usually loaded on the unitary evolution propagator that describes the quantum mechanical unitary dynamics of the quantum system and its ensemble. The highly mixed states could be used in the ensemble quantum computation because the new ensemble quantum computation allows the independence each other between the initial input state and the quantum circuit in a quantum algorithm or sequence. Therefore, the quantum search process based on the new ensemble quantum computation is that the information of the unknown marked state of the search problem is first loaded on the quantum mechanical unitary evolution propagator, then one uses various experiment and measurement techniques such as the NMR multiple-quantum spectroscopy to extract the solution information of the search problem.

2. The simple quantum search sequences on a spin ensemble

First of all, the explicit form of the oracle unitary operation of the quantum search problem is introduced below. In a quantum search algorithm the basic oracle unitary operation $U_f$ is defined as [26]
\[ U_f|x⟩|a⟩ = |x⟩|a \oplus f(x)⟩ \] (1)

where the quantum state \( |x⟩|a⟩ \) is an arbitrary computational basis of the whole quantum system which consists of the work qubits \( I \) and the auxiliary qubits \( S \), the quantum states \( |x⟩ \) and \( |a⟩ \) are the computational bases of the work qubits \( I \) and the auxiliary qubits \( S \), respectively, and the direction sum symbol \( \oplus \) denotes addition modulo 2. The function \( f(x) \) in the quantum search algorithm is defined over the range \( 0 \leq x \leq N - 1 \) \( (N = 2^n) \) by \( f(x) = 1 \) if \( x \) is a solution to the search problem \( (x = s) \), which corresponds to the marked quantum state \( |s⟩ \) of the work qubits \( I \), and \( f(x) = 0 \) if \( x \) is not a solution to the search problem \( (x \neq s) \).

When the basic oracle unitary operation \( U_f \) is applied to the quantum state \( |x⟩|S⟩ \) where the quantum state \( |S⟩ \) of the auxiliary qubits \( S \) is chosen as the superposition: \( |S⟩ = \frac{1}{\sqrt{2}}(|0⟩ - |1⟩) \) it follows from the definition (1) of the oracle unitary operation \( U_f \) that \[ U_f|x⟩|S⟩ = \exp(-i\pi f(x))|x⟩|S⟩ = \begin{cases} -|x⟩|S⟩ & \text{if } x = s \\ |x⟩|S⟩ & \text{if } x \neq s \end{cases} . \] (2)

This shows that only the marked quantum state \( |s⟩ \) is inverted in phase or changes sign of its amplitude but any other state \( |x⟩ \) keeps unchanged when the function values \( f(x) \) are simultaneously evaluated once over all \( x \) values, that is, the oracle unitary operation \( U_f \) is performed once on the quantum system. One can see from Eqs. (1) and (2) that the oracle unitary operation \( U_f \) is independent of any quantum state \( |x⟩ \) of the work qubits \( I \) but related to the quantum state \( |S⟩ \) of the auxiliary qubits \( S \). It must be paid much attention to this point \[17, 18\] when a quantum search algorithm constructed with the oracle unitary operation \( U_f \) and oracle-independent unitary operations is extended to a quantum spin ensemble from its pure-state quantum spin system. For convenient design of a quantum search algorithm or sequence, an auxiliary oracle unitary operation \( U_o \) is introduced below. This oracle unitary operation is expressed as \( U_o(\theta) = U_f^{-1}V_S(\theta)U_f \) \[17\] whose unitary operation on any quantum state of the quantum system is defined by

\[ U_o(\theta)|x⟩|0⟩|1⟩ = U_f^{-1}V_S(\theta)|x⟩|0⟩\oplus f(x)|1⟩ = \exp(-i\theta \delta(f(x), 1))|x⟩|0⟩|1⟩ \] (3)

where the conditional phase-shift unitary operation \( V_S(\theta) \) \[27\] acts only on the auxiliary quantum state \( |S⟩ = |a⟩|b⟩ \) and is defined by \[ V_S(\theta)|a⟩|1⟩ = \exp(-i\theta \delta(a, 1))|a⟩|1⟩ = \begin{cases} \exp(-i\theta)|a⟩|1⟩ & \text{if } a = 1 \\ |a⟩|1⟩ & \text{if } a \neq 1 \end{cases} . \] (4)
Note that the basic oracle unitary operation $U_f$ has the property: $U_f^2 = E$ (unity operation), as can be seen from Eq.(1). Then the oracle unitary operation $U_o$ can also be expressed as $U_o(\theta) = U_f V_S(\theta) U_f$.

The quantum parallelism principle [2] provides the possibility for quantum computation outperforming the classical computation. By the quantum parallelism all the function values $f(x)$ can be evaluated simultaneously over all the input computational base $|x\rangle$ of the quantum system when the oracle unitary operation $U_o$ acts once on any superposition $|\Psi\rangle = \sum_{x=0}^{N-1} a_x |x\rangle |S\rangle$ of the quantum system,

$$U_o(\theta)|\Psi\rangle = \sum_{x=0}^{N-1} \exp(-i\theta \delta(f(x), 1)) a_x |x\rangle |S\rangle$$

$$= \exp(-i\theta a_s |s\rangle |S\rangle) + \sum_{x=0, x \neq s}^{N-1} a_x |x\rangle |S\rangle$$

(5)

where the definition of the function $f(x)$ is used: $f(x) = 1$ if $x = s$; $f(x) = 0$ if $x \neq s$. To analyze conveniently the unitary evolution process of the quantum system and its ensemble during the action of the oracle unitary operation $U_o$, one needs to use the selective phase-shift operation $C_s(\theta)$ [17] to express explicitly the oracle unitary operation. The selective phase-shift operation $C_r(\theta)$ is a diagonal unitary operator which is only applied to the work qubits $I$. It can be expressed in the exponential form

$$C_r(\theta) = \exp(-i\theta D_r)$$

(6)

where the diagonal operator $D_r$ is related to the quantum state $|r\rangle$ of the quantum system and is a LOMSO operator [24], and it is defined as $D_r = diag(0, ..., 0, 1, 0, ..., 0)$, that is, the diagonal element $(D_r)_{rr} = 1$ only for the index $r$ and $(D_r)_{tt} = 0$ for any other index $t \neq r$. It has been proved [17] that when the selective phase-shift operation $C_s(\theta)$ of the marked state $|s\rangle$ acts on any quantum state $|x\rangle |S\rangle$ a phase factor $\exp(-i\theta)$ is generated only when the quantum state $|x\rangle = |s\rangle$, otherwise the quantum state $|x\rangle |S\rangle$ keeps unchanged,

$$C_s(\theta)|x\rangle |S\rangle = \exp(-i\theta \delta_{sx}) |x\rangle |S\rangle.$$  

(7)

When the selective phase-shift operation $C_s(\theta)$ is applied to any superposition $|\Psi\rangle = \sum_{x=0}^{N-1} a_x |x\rangle |S\rangle$ of the quantum system, the unitary evolution process of the quantum system is just the same as Eq.(5) of the oracle unitary operation $U_o(\theta)$,
\[ C_s(\theta)|\Psi\rangle = \exp(-i\theta)a_s|s\rangle|S\rangle + \sum_{x=0,x\neq s}^{N-1} a_x|x\rangle|S\rangle. \]  

(8)

Therefore, the selective phase-shift operation \( C_s(\theta) \) is really equivalent to the oracle unitary operation \( U_o(\theta) \) in the unitary transformation and one can use exactly the selective phase-shift operation \( C_s(\theta) \) to replace the oracle unitary operation \( U_o \) to analyze the unitary evolution process of the quantum system under the action of the oracle unitary operation \( U_o(\theta) \). However, it must be pointed out that the selective phase-shift operation \( C_s(\theta) \) is only applied to the work qubits I and independent of the auxiliary qubits S, while the oracle unitary operation \( U_o(\theta) \) is related to the whole quantum system.

In order to describe conveniently the unitary evolution process of the quantum system under the selective phase-shift operations one may use the quantum state vector, i.e., the unity-number vector \( \{a^s_k = \pm 1\} \) [17] to represent explicitly the selective phase-shift operation \( C_s(\theta) \). In the quantum state vector \( \{a^s_k\} \) representation the diagonal operator \( D_s \) of the marked state can be expressed as

\[ D_s = (1/2 E_1 + a^s_1 I_{1z}) \otimes (1/2 E_2 + a^s_2 I_{2z}) \otimes \cdots \otimes (1/2 E_n + a^s_n I_{nz}). \]  

(9)

It has been shown that the quantum state vector \( \{a^s_k\} \) also determines uniquely the quantum state \( |s\rangle \) [17]. Therefore, one may design a quantum search algorithm or sequence to determine directly the quantum state vector \( \{a^s_k\} \) and then obtains further the marked quantum state \( |s\rangle \) instead of finding directly the marked quantum state \( |s\rangle \) by a quantum search algorithm. This is different from the conventional quantum search strategy [3, 11]. This is also the starting point of the quantum search algorithms or sequences based on the quantum mechanical unitary dynamics. Obviously, the oracle unitary operation \( U_o(\theta) \) contains the desired information to determine certainly the solution \( |s\rangle \) to the quantum search problem. This also can be seen from the fact that the oracle unitary operation \( U_o(\theta) \) is equivalent to the selective phase-shift operation \( C_s(\theta) \), while the latter is determined uniquely by the quantum state vector \( \{a^s_k\} \). Now an NMR ensemble quantum computer operating on the highly mixed states of a spin ensemble is used to solve the quantum search problem. As shown in Eqs.(2), (3), (5), and (8) the oracle unitary operations \( U_f \) and \( U_o(\theta) \) are independent of any quantum state \( |x\rangle \) of the work qubits I but are related to the quantum state \( |S\rangle \) of the auxiliary qubits S. When a quantum search algorithm based on the pure states and built up with the oracle unitary operations and oracle-independent unitary operations is used to determine the marked state \( |s\rangle \) to the search problem
on a spin ensemble, the quantum state $|S\rangle$ of the auxiliary qubits $S$ in the algorithm should be retained or replaced by the effective pure state in the spin ensemble [12], but any mixed states of the work qubits $I$ of the spin ensemble can be used as the input states of the algorithm. For example, for the oracle unitary operation $U_o(\theta)$ the auxiliary quantum state $|S\rangle\langle S|$ ($|S\rangle = |0\rangle|1\rangle$ (see Eq.(3)) in the matrix representation can be expressed as

$$|S\rangle\langle S| = |0\rangle\langle 0| \bigotimes_k |1\rangle\langle 1| = \frac{1}{2}E + \frac{1}{2}(S_{1z} - S_{2z}) - S_{1z}S_{2z}. \quad (10)$$

Then the auxiliary quantum state $|S\rangle\langle S|$ can be replaced by the effective pure state taking the same form as Eq.(10) and the initial quantum state $|x\rangle$ of the work qubits $I$ of the algorithm can be replaced even by any highly mixed state of the work qubits $I$ in the spin ensemble. However, the mixed state must be related to the marked state $|s\rangle$ in order that the quantum state vector $\{a_k^s\}$ of the marked state $|s\rangle$ can be determined certainly by the algorithm. Note that any magnetization operator $I_{k\mu}$ ($k = 1, 2, ..., n; \mu = x, y$) and the diagonal operator $D_s$ of the marked state do not commute. It is easy to prove that the following density operators $\rho(0)$ of the spin ensemble always contains the marked state $|s\rangle$,

$$\rho(0) = \rho_I(0)|S\rangle\langle S| = (\alpha E + \varepsilon_k I_{k\mu})|S\rangle\langle S| \quad (k = 1, 2, ..., n; \mu = x, y), \quad (11a)$$

and

$$\rho(0) = \rho_I(0)|S\rangle\langle S| = (\alpha E + \sum_{k=1}^{n} \varepsilon_k I_{k\mu})|S\rangle\langle S| \quad (11b)$$

where the operator $E$ is unity operator, $\varepsilon_k$ is the spin polarization parameter of the spin $k$ in the spin ensemble, and $\alpha$ is the normalized constant.

Now one can analyze conveniently the time evolution process of a quantum spin system or its spin ensemble under the action of the oracle unitary un itary describes the action of the selective phase-shift operation $C_s(\theta)$ on any density operator $\rho_I(0)$ of the work qubits $I$ of the spin ensemble [28]:

$$C_s(\theta)\rho_I(0)C_s(\theta)^{-1} = \rho_I(0) - (1 - \cos\theta)[\rho_I(0), D_s] + i\sin\theta[\rho_I(0), D_s] + [(1 - \cos\theta)^2 + \sin^2\theta]D_s\rho_I(0)D_s \quad (12)$$

where the commutation $[\rho_I(0), D_{s\mu}] = \rho_I(0)D_{s\mu} + D_{s\mu}\rho_I(0)$ and $[\rho_I(0), D_s] = \rho_I(0)D_s - D_s\rho_I(0)$. A general unitary transformation with a sequence of many selective phase-shift operations $\{C_k(\theta_k)\}$ can be derived directly from the basis unitary transformation (12),

$$U_o(\theta_0, \theta_1, ..., \theta_{m-1})\rho_I(t)U_o(\theta_0, \theta_1, ..., \theta_{m-1})^{-1} = \rho_I(t) - [\rho_I(t), \sum_{k=0}^{m-1} (1 - \cos\theta_k)D_k] + i[\rho_I(t), \sum_{k=0}^{m-1} D_k \sin\theta_k]$$
is applied to all the spins of the work qubits $I$.

The density operator of Eq.(14) is reduced to the form to cancel all the zero-quantum coherence. In the final density operator

and then a zero-quantum dephasing pulse \cite{29} is applied to the spin ensemble

order multiple-quantum coherence including the single-quantum coherence, a magnetic field \cite{6, 7} is applied to the spin ensemble to cancel all the non-zero-selective phase-shift operations:

\[ U_o(\theta_0, \theta_1, ..., \theta_{m-1}) = \prod_{k=0}^{m-1} C_k(\theta_k). \]

On the basis of the unitary transformation (12) one can design a simple quantum search sequence to solve efficiently the quantum search problem on a spin ensemble. First, the spin ensemble is prepared at the initial mixed state of Eq.(11b). Then the oracle unitary operation $U_o(\theta)$ is applied to the spin ensemble. The spin ensemble will evolve according to the unitary transformation (12). This unitary transformation is given explicitly below by inserting the diagonal operator $D_o$ of Eq.(9) in the quantum state vector $\{a^s_k\}$ representation and the initial density operator of Eq.(11b) with the index $\mu = y$ into Eq.(12),

\[
U_o(\theta) \sum_{k=1}^{n} \varepsilon_k I_{ky} U_o(\theta)^{-1} = \sum_{k=1}^{n} \varepsilon_k I_{ky} - (1 - \cos \theta) \sum_{k=1}^{n} \left( \frac{1}{2} E_1 + a^s_{11} I_{1z} \right) \bigotimes ... \bigotimes \left( E_{k-1} + a^s_{k-1} I_{k-1z} \right) \bigotimes \left( E_{k} + a^s_{k} I_{kz} \right) - \sin \theta \sum_{k=1}^{n} \left( \frac{1}{2} E_1 + a^s_{11} I_{1z} \right) \bigotimes ... \bigotimes \left( \frac{1}{2} E_{k-1} + a^s_{k-1} I_{k-1z} \right) \bigotimes \left( \varepsilon_k a^s_{k} I_{kz} \right) \bigotimes \left( \frac{1}{2} E_{k+1} + a^s_{k+1} I_{k+1z} \right) \bigotimes ... \bigotimes \left( \frac{1}{2} E_n + a^s_{n} I_{nz} \right) \]  

(14)

where the term proportional to the unity operator $E$ and the auxiliary quantum state $|S\rangle \langle S|$ of the initial density operator of Eq.(11b) are omitted without losing generality. Note that here the auxiliary quantum state $|S\rangle \langle S|$ is taken as the effective pure state and is of the LOMSO operator subspace, and hence it will keep unchanged when a $z-$direction gradient magnetic field is applied to the spin ensemble \cite{6, 7}. Obviously, the generated mixed state of Eq.(14) under the action of the oracle unitary operation $U_o(\theta)$ on the spin ensemble is quite complicated, but it can be simplified by using the selective decoherence manipulation. First, a 90 degree hard pulse along $y-$direction is applied to all the spins of the work qubits $I$. Then a $z-$direction gradient magnetic field \cite{6, 7} is applied to the spin ensemble to cancel all the nonzero-order multiple-quantum coherence including the single-quantum coherence, and then a zero-quantum dephasing pulse \cite{29} is applied to the spin ensemble to cancel all the zero-quantum coherence. In the final density operator of Eq.(14) is reduced to the form
\[ U_o(\theta) \sum_{k=1}^{n} \varepsilon_k I_{ky} U_o(\theta)^{-1} \Rightarrow \rho_f = (2/N) \sum_{k=1}^{n} \varepsilon_k a_k^* I_{kz} \]  

(15)

Obviously, the density operator \( \rho_f \) of Eq.(15) carries the desired information, i.e., the quantum state vector \( \{a_k^*\} \), to solve the quantum search problem. Now, a read pulse \( 90^\circ-x \) is applied to all the spins of the work qubits \( I \) and then the NMR receiver is turned on to record in phase-sensitive mode the NMR signal of the density operator \( \rho_f \) of Eq.(15). In order to cancel the effect of the auxiliary qubits \( S \) of Eq.(10) on the recorded NMR signal the spins of the auxiliary qubits \( S \) are decoupled during recording the NMR signal. From the fast Fourier transform spectrum of the recorded NMR signal one can determine certainly the quantum state vector \( \{a_k^*\} \) when the signal-to-noise ratio of the NMR signal of Eq.(15) is high enough. The density operator of Eq.(15) shows that the NMR signal intensity is proportional to the factor \( (2/N) \), indicating that the NMR signal intensity reduces exponentially as the qubit number of the work qubits \( I \). This characteristic feature is similar to the classical NMR quantum computation based on the effective pure states [15]. Therefore, the above quantum search algorithm can only be used to solve efficiently the quantum search problem with a few qubits.

However, the significant difference between the quantum search algorithms based on the spin ensemble and the pure quantum state [3] or the effective pure state [11] is that the Grover quantum search algorithm based on the pure quantum states [3] or the effective pure state [11] can achieve only quadratic speedup over the classical counterparts due to the limit of quantum mechanical measurement [6], which is a limit in quantum mechanical principle, that is, the conventional Grover algorithm is a quadratic but not polynomial-time algorithm, but the ensemble quantum search sequence can achieve an exponential-time speedup over the classical counterparts, that is, this sequence is a polynomial-time one (one performs only twice times the oracle unitary operation \( U_f \) in the algorithm) and really moves the frontier between the solvability and intractability of complexity problems. Though the final output NMR signal of the ensemble quantum search sequence reduces exponentially as the size of the solved search problem, this limit is in techniques but not in quantum mechanical principles and therefore, this limit could be overcome as the relevant techniques are improved [7, 8, 22, 23, 24]. There is also another advantage for the ensemble quantum search sequence that the sequence can be run even on a spin system with a short decoherence time since in the whole sequence the oracle unitary operation \( U_f \) is performed only twice times.
There may be two possible schemes [18] to improve the above ensemble quantum search sequence so that it could be used to solve the search problem with a larger number of qubits. One possible scheme is that the initial spin polarization of a spin ensemble is enhanced sufficiently with various possible NMR signal enhancement techniques including spin polarization transfer techniques [6, 7, 22] and other techniques [23, 24] so that the NMR signal of the density operator of Eq.(15) still can be observable even in the spin system with a larger number of qubits, although the final NMR signal of the sequence is reduced exponentially as the qubit number of the spin system. This scheme retains the original advantage of the sequence that it still can be run on the spin systems with a short decoherence time. However, this scheme is rigorous in technique. Another scheme is that one could enhance the final detectable NMR signal by running a quantum search sequence that consists of a number of the oracle unitary operations and oracle-independent unitary operations on the spin ensemble. But this scheme requires that the spin ensemble must have a long decoherence time. Obviously, there is a maximum call number $M_s$ of the oracle unitary operation $U_o(\theta)$ or a maximum running time $T_{2\text{max}}$ of the quantum search sequence in a spin ensemble due to the decoherence effect. The final output NMR signal of the sequence will decrease simply but not be enhanced again as increasing the call number of the oracle unitary operation in the sequence if the call number is larger than the maximum one $M_s$. This is because in that case running the sequence will take more time than $T_{2\text{max}}$, so that the decoherence effect becomes the dominant effect negatively upon the final NMR signal.

A simple method to enhance the output NMR signal by many calls of the oracle unitary operation is the conventional spin echo experiments [5, 7]. The qubit interaction in the diagonal operator $D_s$ of the marked state $|s\rangle$ can be decoupled one by one by the spin-echo experiments. First, any Hamiltonian $D_s(n - k)$ ($k = 1, 2, \ldots, n - 1$) that is independent of $k$ qubits of the spin system is prepared with the spin echo sequences by starting the diagonal operator $D_s$,

$$D_s(n - 1) = D_s + \exp(-i\pi I_{nx})D_s \exp(i\pi I_{nx}),$$

$$D_s(n - 2) = D_s(n - 1) + \exp(-i\pi I_{n-1x})D_s(n - 1) \exp(i\pi I_{n-1x}), \ldots,$$

$$D_s(n - k) = D_s(n - k + 1) + \exp(-i\pi I_{n-k+1x})D_s(n - k + 1) \exp(i\pi I_{n-k+1x}),$$

where $D_s(n - k) \equiv \bigotimes_{l=1}^{n-k} (\frac{1}{2} E_l + a_l^z I_{lz})$. Then one builds up a new auxiliary oracle unitary operation $V_{os}(k, \theta) = \exp[-i\theta D_s(n - k)]$ with the Hamilto-
nian $D_s(n - k)$. There are $2^k$ oracle unitary operations $U_o(\theta)$ in the unitary transformation $V_{os}(k, \theta)$. Now using the oracle unitary operation $V_{os}(k, \theta)$ to replace the oracle unitary operation $U_o(\theta)$ in Eq.(14) to perform quantum search one can find that the final NMR signal intensity of Eq.(15) is proportional to the factor $2^k$, indicating that the output NMR signal intensity of the new quantum search sequence increases linearly as the call number of the oracle unitary operation. Obviously, this is an ineffective sequence to extend the realizable size of the NMR ensemble quantum computation. An improved approach to the simple spin-echo experiments is that following the action of the unitary operation $V_{os}(k, \theta)$ on the initial density operator one applies a sequence of non-oracle selective unitary operations $\prod_r C_r(\theta)$ to the spin ensemble. Then the NMR signal intensity of Eq.(15) is proportional to the sum $\sum_k \varepsilon_k (\sum_s a_s^k + \sum_r a_r^k)I_{kx}$, where the quantum state vectors $\{a_s^k\}$ and $\{a_r^k\}$ are related to the oracle and the non-oracle selective unitary operations, respectively. Now, if the factors $\sum_s a_s^k$ and $\sum_r a_r^k$ have the same sign then the final NMR signal is enhanced, otherwise it is reduced. Therefore, the NMR signal intensity may be enhanced when the non-oracle selective unitary operations are chosen properly. The more the number of the non-oracle selective unitary operations is in the sequence, the stronger the NMR signal is. However, there is a question that how many non-oracle selective unitary operations can be used to enhance effectively the NMR signal under the condition that the quantum state vector $\{a_s^k\}$ can be extracted certainly from the final NMR signal intensity. This question limits really the maximum enhancement of the NMR signal by the sequence since the more the non-oracle selective unitary operations are used in the sequence it becomes more difficult for the solution information of the search problem to be extracted from the NMR signal. In order to improve further the method the NMR multiple-quantum spectroscopy is proposed to solve the search problem in the following. I will discuss the quantum search NMR multiple-quantum spectroscopy from the viewpoint of the NMR experiments and leave its complexity problem in the future.

3. The NMR multiple-quantum spectroscopy

The basic idea for the multiple-quantum NMR spectroscopic method used
to perform the quantum search is that information of the unknown marked state \( |s \rangle \) is first loaded on the multiple-quantum coherence of a spin ensemble by using iterative sequences of the oracle unitary operations \( U_o(\theta) \) and oracle-independent unitary operations, the search problem then could be solved by extracting the desired information of the marked state from these NMR multiple-quantum transition spectra obtained by NMR measurement techniques. The important point for the method is that the multiple-quantum coherence of the spin ensemble need first to be excited effectively by a sequence of the oracle unitary operations and oracle-independent unitary operations. Here a general (formal) quantum search sequence based on the multiple-quantum NMR spectroscopy [7, 8] is proposed. The sequence is constructed with the oracle unitary operations and oracle-independent unitary operations and may be divided into the following several steps:

(a) The suitable initial input state for the quantum search sequence is prepared at the mixed state of a spin ensemble such as Eq.(11a) or (11b),

\[
\rho(0) = \rho_I(0) |S\rangle \langle S| = (\alpha_0 E + \sum_{k=1}^{n} \epsilon_k I_{kp}) |S\rangle \langle S| (p = x, y, z).
\]

(b) Apply the iterative sequence \( U(D_s) \) which is built up with the oracle unitary operations \( U_o(\theta) \) and oracle-independent unitary operations to the initial mixed state \( \rho(0) \) to excite the multiple-quantum coherence,

\[
\rho(\tau_1) = U(D_s) \rho(0) U(D_s)^+.
\]

(c) During the \( t_1 \) evolution period under the effective spin Hamiltonian \( H \) the multiple-quantum coherence are labelled by their own frequencies so that various order multiple-quantum coherence can be distinguished clearly in NMR multiple-quantum spectra,

\[
\rho(t_1, \tau_1) = \exp(-iHt_1) \rho(\tau_1) \exp(iHt_1).
\]

(d) Apply the second iterative sequence \( V(D_s) \) of the oracle unitary operations and oracle-independent unitary operations to convert the multiple-quantum coherence into the single-quantum coherence so that they can be detected by NMR probes.

\[
\rho(\tau_2, t_1, \tau_1) = V(D_s) \rho(t_1, \tau_1) V(D_s)^+.
\]

(e) The NMR signal detection along with \( q - \)direction \( (q = x, y, z) \)

\[
\rho_f = \rho_f(t_1) = Tr \{ \sum_{k=1}^{n} I_{kq} \rho(\tau_2, t_1, \tau_1) \}.
\]

The longitudinal component \( I_{kz} \) in the final NMR signal \( \rho_f \) is not observable in NMR experiments, but it becomes observable if an additional 90 degree pulse converts it into the single-quantum coherence. The multiple-quantum coherence are usually detected indirectly by NMR measurement techniques
such as the two-dimensional NMR method [7, 8]. The usual two-dimensional NMR experiment methods have been developed to do quantum computation on the basis of the concept of the effective pure states [12], but here NMR multiple-quantum spectroscopy is emphasized. The oracle iterative sequence $U(D_s)$ carries the information of the marked state $|s\rangle$ and in the step (b) the information is really transferred into the multiple-quantum coherence of the spin ensemble through applying the iterative sequence $U(D_s)$ to the initial density operator of the spin ensemble. The multiple-quantum coherence (or transitions) are detected indirectly by the two-dimensional NMR measurement techniques [7, 8], which is achieved by the steps (c), (d) and (e) in the sequence. Obviously, the output NMR signal $\rho_f(t_1)$ carries the information of the marked state $|s\rangle$. Then by fast Fourier transforming the output NMR signal $\rho_f(t_1)$ on the time variable $t_1$ one obtains the multiple-quantum spectrum. Now the spectral parameters including the multiple-quantum resonance frequencies, phase, and intensities of the multiple-quantum spectrum contain the information of the marked state $|s\rangle$. Then one could extract the quantum state vector $\{a_s^k\}$ and hence find the marked state $|s\rangle$ from these spectral parameters by NMR data processing.

3.1 The expression for the output NMR signal

For convenient discussion, here consider a homonuclear spin ensemble where the spin polarization parameter $\varepsilon_k$ for all the spins in the ensemble are the same ($\varepsilon = \varepsilon_k$). Then the initial density operator of work qubits $I$ in the step (a) is written briefly as $\rho_I(0) = \varepsilon F_p = \varepsilon \sum_{k=1}^{n} I_{kp}$ without losing generality since the auxiliary quantum state $|S\rangle\langle S|$ the term proportional to the unity operator $E$, and the spin polarization factor $\varepsilon_k$ keep unchanged during the evolution process of the quantum search sequence. From the steps (a)-(e) of the sequence the final NMR signal intensity of the sequence can be generally expressed as

$$
\rho_f(t_1) = \text{Tr}\{F_q V(D_s) \exp(-iHt_1) U(D_s) \varepsilon F_p U(D_s)^+ \exp(iHt_1) V(D_s)^+\} \\
= \varepsilon \text{Tr}\{V(D_s)^+ F_q V(D_s) \exp(-iHt_1) U(D_s) F_p U(D_s)^+ \exp(iHt_1)\} \\
$$

(16)

where the invariance of the trace operation $\text{Tr}$ to cyclic permutation has been introduced. In high-resolution NMR spectroscopy the spin Hamiltonian $H$ of the spin system in the spin ensemble in a high magnetic field is a longitudinal magnetization and spin order ($LOMSO$) operator and usually can be written as [7, 8]
\[ H = \sum_{k=1}^{n} \Omega_k I_{kz} + \sum_{k>l}^{n} 2\pi J_{kl} I_{kz} I_{lz} \]  

With the help of the eigenbase \( |k\rangle \) of the Hamiltonian \( H \), \( H|k\rangle = \omega_k|k\rangle \) \((\hbar = 1)\), which are also the usual quantum computational bases, the NMR signal of Eq.(16) can be expanded as

\[ \rho_f(t_1) = \varepsilon \sum_{j,k} Q^{\ast}_{jk} P_{jk} \exp(-i\omega_{jk}t_1) \]  

where the multiple-quantum transition frequency \( \omega_{jk} = \omega_j - \omega_k \) and the Hermitian operators \( P(D_s) \) and \( Q(D_s) \) are defined as

\[ P(D_s) = U(D_s) F_P U(D_s)^+, \]  

\[ Q(D_s)^+ = Q(D_s) = V(D_s)^+ F_Q V(D_s), \]  

and the matrix elements \( P_{jk} \) and \( Q_{jk} \) are given by

\[ P_{jk} = \langle j | P(D_s) | k \rangle, \quad Q_{jk} = \langle j | Q(D_s) | k \rangle. \]

Although the eigenvalue \( \omega_k \) of the Hamiltonian \( H \) is independent of the quantum state vector \( \{a^s_k\} \) of the marked state \( |s\rangle \), the excited multiple-quantum transition frequencies \( \{\omega_{jk}\} \) may contain the information of the marked state \( |s\rangle \) besides the amplitudes and phases of the multiple-quantum transitions since the excitation of the multiple-quantum coherence by the oracle unitary sequence \( U(D_s) \) is dependent on the marked state. Actually, the oracle unitary sequence \( U(D_s) \) decides which orders multiple-quantum coherence are excited effectively and finally detected in the quantum search sequence. If the inphase multiple-quantum spectrum is required to be observed for a given quantum order one had better require that the oracle unitary sequence \( V(D_s) \) satisfies the relation [30]:

\[ Q(D_s)^+ = \exp(-i\varphi F_z) P(D_s) \exp(i\varphi F_z). \]  

Then the NMR signal intensity of Eq.(18) can be further expressed as

\[ \rho_f(t_1) = \varepsilon \sum_{m} \sum_{j,k} |P_{jk}|^2 \exp(i m \varphi) \exp(-i\omega_{jk}t_1) \]  

where the quantum order \( m = M_j - M_k \) and \( M_k \) is the magnetic quantum number: \( F_z|k\rangle = M_k|k\rangle \) \((\hbar = 1)\). The NMR signal of Eq.(21) indicates that each \( m \)-order multiple-quantum transition has the same phase, although the multiple-quantum transitions with different quantum order \( m \) may have a phase difference. Therefore, by using separation techniques for different order multiple-quantum transitions such as TPPI technique [7, 8] one can record the inphase multiple-quantum peaks with their own resonance frequencies or quantum order in the multiple-quantum spectrum. The intensities, frequencies, and phases of the multiple-quantum peaks carry the information of the marked state. Then from these spectral parameters one could...
extract the desired information of the marked state. Although the inphase multiple-quantum spectrum of Eq.(21) may be more convenient and useful in the measurement of structural properties of molecules in NMR multiple-quantum spectroscopy [30], the multiple-quantum spectrum of Eq.(18) may contain richer information of the marked state and may be more useful to solve the quantum search problem.

If the effective spin Hamiltonian is prepared as $H = \omega F_z$ instead of that one of Eq.(17) in the $t_1$ evolution period, all the multiple-quantum coherence with the same quantum order have the same precession frequency. That is, for all the $k-$order multiple-quantum coherence their precession frequencies are the same and equal $f_k = k\omega$ ($k = 0, \pm 1, \pm 2, ..., \pm n$). In this case there are $(2n + 1)$ different order multiple-quantum resonance peaks with their own resonance frequencies $\{f_k\}$ in the multiple-quantum spectrum for an $n-$qubit spin system. Since different order multiple-quantum coherence in the density operator $\rho_f(t_1)$ of Eq.(18) are labelled with their own transition frequencies (or precession frequencies), they can be distinguished in the NMR multiple-quantum frequency spectrum. It is known that number of independent multiple-quantum coherence increases exponentially as the qubit number in a spin system [7]. If each multiple-quantum coherence has its own precession frequency in a spin system, then amplitude of each multiple-quantum coherence will decrease exponentially as the qubit number and this will make each multiple-quantum spectral peak unobservable in the multiple-quantum spectrum of a spin system with many qubits since the total amplitude of the multiple-quantum coherence of a spin system is fixed and is determined by the initial density operator. The importance to choose the spin Hamiltonian $H = \omega F_z$ to label the multiple-quantum coherence in the $t_1$ evolution period instead of the spin Hamiltonian of Eq.(17) therefore becomes very clear: all the same order multiple-quantum coherence have the same precession frequency and their resonance frequency peaks in the multiple-quantum spectrum can be coherently co-added in amplitude and become a single spectral peak, as can be seen in the NMR signal of Eq.(18), so that the multiple-quantum spectral peak may increase greatly in a favorable case. This shows that at least some lower order multiple-quantum spectral peaks in the spin ensemble do not reduce exponentially as the qubit number if the initial density operator is excited effectively to the multiple-quantum coherence by the oracle iterative sequence $U(D_s)$. Therefore, complexity of the quantum search sequence now is only dependent on the excitation of the multiple-quantum coherence by the oracle iterative sequence $U(D_s)$ and the
extraction of the solution information of the search problem by the NMR data processing. Of course, it could become more difficult to extract the information of solution of the search problem using this method labelling multiple-quantum coherence with respect to that one using the Hamiltonian of Eq.(17). By the NMR signal expression (18) one can explicitly calculate the output NMR signal intensities of various order multiple-quantum transitions,

$$\rho_f(t_1) = \sum_{m=-n}^{n} \left[ \varepsilon \sum_{j,k}' Q_{jk}^* P_{jk} \right] \exp(-im\omega t_1) \quad (22)$$

where the sum \(\sum'\) runs only those indexes \((j, k)\) with the quantum order \(m = M_j - M_k\). The \(m\)–order multiple-quantum peak intensity and phase therefore are determined from

$$I(m) = \left[ \varepsilon \sum_{j,k}' Q_{jk}^* P_{jk} \right]. \quad (23)$$

Now the information of solution of the search problem could be extracted from the spectral parameters \(I(m)\) by NMR data processing.

### 3.2 The multiple-quantum operator algebra spaces and the construction of the oracle unitary sequences

The multiple-quantum operator algebra space \([24, 25]\) states that the whole Liouville operator space of a spin ensemble contains the even-order multiple-quantum operator subspace, the zero-quantum operator subspace, and the longitudinal magnetization and spin order (LOMSO) operator subspace and each former subspace contains the followed subspaces. For example, the zero-quantum operator subspace contains the whole LOMSO operator subspace. There are important properties for the zero-quantum operators and the even-order multiple-quantum operators. Any operator of the Liouville operator space will keep its quantum order unchanged when it is acted on by a zero-quantum operator and the parity of quantum order of an operator of the Liouville operator space can not be changed when the operator is acted on by an even-order multiple-quantum operator \([24, 31]\). With the help of the closed properties of these operator subspaces one may simplify the construction of the oracle unitary sequences \(U(D_{s})\) and \(V(D_{s})\). In general, the oracle unitary sequences \(U(D_{s})\) and \(V(D_{s})\) are composed of the oracle unitary operation \(U_\alpha(\theta)\) and oracle-independent unitary operations. Suppose that the oracle unitary sequence \(U(D_{s})\) is constructed in the form:

$$U(D_{s}) = R_{k}O_{k-1}R_{k-1}\ldots O_{1}R_{1} \quad (24)$$

where each unitary operation \(R_{k}\) is an oracle-independent unitary operation
and $O_k = O_k(D_s)$ an oracle unitary operation sequence which contains the oracle unitary operation $U_o(\theta)$. Now assume that the initial mixed state $\rho_I(0)$ in the quantum search sequence is taken as the longitudinal magnetization operator, $\rho_I(0) = \sum_{k=1}^{n} \varepsilon_k I_{kz}$, which is also a zero-quantum operator. There may be two simpler schemes to load the information of the marked state $|s\rangle$ on the multiple-quantum spectrum. One scheme is that only the zero-quantum transitions are used to carry the information of the marked state. The zero-quantum operator subspace is the smaller operator subspace in the Liouville operator space. Then it is a better scheme to load the information of the marked state on the zero-quantum operator subspace since this could simplify the loading and extraction of the desired information of the marked state. Obviously, this requires that the oracle unitary sequence $U(D_s)$ should be prepared as a zero-quantum unitary operator. When the oracle unitary sequence $U(D_s)$ is applied to the initial mixed state $\rho_I(0)$ only the zero-quantum coherence of the spin system can be excited according to closed property of the multiple-quantum operator algebra space since the initial mixed state $\rho_I(0)$ is also a zero-quantum operator. In order to distinguish the effects between the oracle unitary operations $\{O_k\}$ and the oracle-independent unitary operations $\{R_k\}$ on the excitation of the zero-quantum coherence it could be better to choose the unitary operations $\{R_k\}$ as the LOMSO unitary operators and the oracle unitary operations $\{O_k\}$ as the zero-quantum unitary operators. It is known from multiple-quantum operator algebra space formalism that any pair of the LOMSO operators commute with each other, and the closed property of the zero-quantum operator subspace shows that any zero quantum operator is transformed unitarily into a zero-quantum operator when it is acted on by any zero-quantum unitary operator. Then the zero-quantum coherence will be excited only by the oracle unitary operations $\{O_k\}$, while the LOMSO unitary operations $\{R_k\}$ should have not net contribution to the excitation of the zero-quantum coherence. But the unitary operators $\{R_k\}$ could be chosen properly so that the excitation efficiency of the zero-quantum coherence can be enhanced. If the zero-quantum coherence spectra are observed in the output NMR signal of the quantum search sequence it can be sure that the zero-quantum spectra contain the information of the marked state. Another scheme is that the nonzero order multiple-quantum coherence and especially the even-order multiple-quantum coherence are used to carry the information of the marked state. According to the multiple-quantum operator algebra space formalism
all the even-order multiple-quantum operators form a closed operator sub-
space of the Liouville operator space. The closed property of the even order
multiple-quantum operator subspace shows that the initial density operator
(a $LOMSO$ operator) will be transferred into even order multiple-quantum
coherence if it is acted on by an even-order multiple-quantum unitary oper-
ator. Therefore, if all the oracle unitary operations $\{O_k\}$ are taken as the
even-order multiple-quantum unitary operators and the oracle-independent
unitary operations $\{R_k\}$ as the proper $LOMSO$ or zero-quantum unitary
operators, then even order multiple-quantum coherence will be excited by
the oracle unitary sequence $U(D_s)$ in the spin ensemble and moreover, the
nonzero even-order multiple-quantum coherence can only be excited by the
oracle unitary operations $\{O_k\}$ and the oracle-independent unitary opera-
tions $\{R_k\}$ will have not net contribution to the excitation of the nonzero-
order multiple-quantum coherence when the oracle unitary sequence $U(D_s)$
is applied to the initial density operator $\rho_I(0)$. Therefore, the information
of the marked state now is contained mainly in the nonzero even order multiple-
quantum spectra of the output NMR signal of the sequence. How easy to ex-
tact the information $\{a_k^s\}$ of the marked state from these multiple-quantum
spectra? This depends on what the explicit forms of the oracle-independent
operators $\{R_k\}$ and the oracle unitary operations $\{O_k\}$ are taken. The im-
portance is that the oracle unitary sequence $U(D_s)$ needs to be constructed
properly so that the multiple-quantum coherence can be excited effectively.

(a) the preparation for the zero-quantum unitary operators

There are a number of methods to construct the zero-quantum unitary
operations with a sequence of the oracle unitary operations $U_o(\theta)$ and oracle-
independent unitary operations. One simple and systematic method is that
the preparation can be performed by the discrete Fourier analysis and the
phase cycling technique [17] by starting the diagonal operator $D_s$ of the
marked state $|s\rangle$. According to the multiple-quantum operator algebra space
any diagonal operator $D_l$, including the diagonal operator $D_s$ of the marked state $|s\rangle$, can be expressed as a linear combination of the $LOMSO$ basis
product operators $\{Z_k\} = \{E, I_{kz}, 2I_{kz}I_{lz}, 4I_{kz}I_{lz}I_{mz}, \ldots, 2^{n-1}I_{1z}I_{2z}...I_{nz}\}$ including the unity operator $E$ ($E = Z_0$) [24],
$$D_k = \sum_{l=0}^{N-1} (A)_{kl} Z_l.$$  \hspace{1cm} \text{(25)}

Then the Hermitian product operators $X_l = \exp(-i\frac{\pi}{2}F_y)Z_l \exp(i\frac{\pi}{2}F_y)$ can be
constructed by
\[
X_l = 2^l-1 I_{k_1} I_{k_2} \ldots I_{k_1} = \exp(-i \frac{\pi}{2} F_y) [\sum_{j=0}^{N-1} (A^{-1})_{ij} D_j] \exp(i \frac{\pi}{2} F_y) \tag{26}
\]
and in a more general case any linear operator function \(f(D_s)\) related to the diagonal operator \(D_s\) of the marked state \(|s\rangle\) and any other diagonal operators \(D_t\) by
\[
f(D_s) = \sum_{l=1} b_l X_l
\]
\[
= \exp(-i \frac{\pi}{2} F_y) [\sum_{l=1} b_l \sum_{j=0}^{N-1} (A^{-1})_{ij} D_j] \exp(i \frac{\pi}{2} F_y) \tag{27}
\]
where \(b_l\) is a coefficient. Now by starting the operator function of Eq.(27) one can prepare the Hermitian zero-quantum operators with aid of the discrete Fourier analysis and the phase cycling technique [17],
\[
H_{zq}(D_s) = \frac{1}{N_1} \sum_{k=0}^{N_1-1} \exp(-i \varphi_k F_z) f(D_s) \exp(-i \varphi_k F_z)
\]
\[
= \frac{1}{N_1} \sum_{l h=1}^n J_{hl} \{a^s_k\} (I^+_l I^-_h + I^+_h I^-_l) + \frac{1}{N_1} \sum_{q>p>h>l=1} J_{klpq} \{a^s_k\} (I^+_l I^-_h I^-_p + I^-_h I^-_h I^+_p + I^-_h I^-_h I^-_p)
\]
\[
+ J_{klpq} \{a^s_k\} (I^+_l I^-_h I^-_p I^-_q + I^-_h I^-_h I^+_p I^-_q) + \ldots, \tag{28}
\]
where the phase \(\varphi_k = k2\pi/N_1, k = 0, 1, 2, \ldots, N_1 - 1\) and the integer \(N_1 \geq n\). By the \(N_1\)-step phase phase cycling systematically only the zero-quantum operator \(H_{zq}(D_s)\) of the operator function \(f(D_s)\) is retained but all nonzero order multiple-quantum operators are cancelled. The zero-quantum unitary operator then can be constructed generally with the Hermitian zero-quantum operator of Eq.(28),
\[
U_{zq}(D_s, t) = \exp[-i H_{zq}(D_s)t]. \tag{29}
\]
Since the operator function \(f(D_s)\) contains the diagonal operator \(D_s\) of the marked state \(|s\rangle\) the zero-quantum operator \(H_{zq}(D_s)\) of Eq.(28) carries the information of the marked state \(|s\rangle\). The zero-quantum unitary operator of Eq.(29) may be further decomposed into a sequence of the oracle unitary operations and oracle-independent unitary operations by using the Trotter-Suzuki formula [32, 33]:
\[
\lim_{m \to \infty} (e^{-iA_1/m} e^{-iA_2/m} \ldots e^{-iA_n/m})^m = e^{-i(A_1+A_2+\ldots+A_n)}. \tag{30}
\]
Now the decomposition (30) of the zero-quantum unitary operator of Eq.(29) may be used as the oracle unitary sequence \(U(D_s)\) to excite the zero-quantum
coherence in the quantum search sequence.

**(b) the preparation for the nonzero-order multiple-quantum unitary operators**

Using the discrete Fourier analysis and the phase cycling technique [17] by starting the diagonal operator $D_s$ of the marked state $|s\rangle$ one still can construct generally any nonzero order multiple-quantum coherence operators and further uses the operators to construct the multiple-quantum unitary operators. But a simple scheme to prepare a nonzero-order multiple-quantum unitary operator as a sequence of the oracle unitary operations and oracle-independent unitary operations is given below. For any Hermitian product operator $X_l = 2^{l-1}I_{k_1}I_{k_2}...I_{k_l}$ of the $x-$direction product operator set $\{E, \ E_{k_1}, 2I_{k_1}I_{k_2}, 4I_{k_1}I_{k_2}I_{k_3}, ...\}$ one may construct a number of the nonzero order multiple-quantum operators such as $i[X_l, (D_0 \pm D_{N-1})]_\pm$. The typical two of which are given explicitly by

$$i[X_l, (D_0 - D_{N-1})] = +\frac{1}{2} \bigotimes_{j=1}^l (I_{k_j} - iI_{k_j}) \bigotimes \left( \frac{1}{2} I_{k_{j+1}} + I_{k_{j+1}} \right) \bigotimes \left( \frac{1}{2} I_{k_n} + I_{k_n} \right)$$

$$-\frac{1}{2} \bigotimes_{j=1}^l (I_{k_j} + iI_{k_j}) \bigotimes \left( \frac{1}{2} I_{k_{j+1}} - I_{k_{j+1}} \right) \bigotimes \left( \frac{1}{2} I_{k_n} - I_{k_n} \right)$$

$$-\frac{1}{2} \bigotimes_{j=1}^l (I_{k_j} + iI_{k_j}) \bigotimes \left( \frac{1}{2} I_{k_{j+1}} + I_{k_{j+1}} \right) \bigotimes \left( \frac{1}{2} I_{k_n} + I_{k_n} \right)$$

$$+\frac{1}{2} \bigotimes_{j=1}^l (I_{k_j} - iI_{k_j}) \bigotimes \left( \frac{1}{2} I_{k_{j+1}} - I_{k_{j+1}} \right) \bigotimes \left( \frac{1}{2} I_{k_n} - I_{k_n} \right), \quad (31)$$

$$[X_l, (D_0 + D_{N-1})] = +\frac{1}{2} \bigotimes_{j=1}^l (I_{k_j} - iI_{k_j}) \bigotimes \left( \frac{1}{2} I_{k_{j+1}} + I_{k_{j+1}} \right) \bigotimes \left( \frac{1}{2} I_{k_n} + I_{k_n} \right)$$

$$+\frac{1}{2} \bigotimes_{j=1}^l (I_{k_j} + iI_{k_j}) \bigotimes \left( \frac{1}{2} I_{k_{j+1}} - I_{k_{j+1}} \right) \bigotimes \left( \frac{1}{2} I_{k_n} - I_{k_n} \right)$$

$$+\frac{1}{2} \bigotimes_{j=1}^l (I_{k_j} + iI_{k_j}) \bigotimes \left( \frac{1}{2} I_{k_{j+1}} + I_{k_{j+1}} \right) \bigotimes \left( \frac{1}{2} I_{k_n} + I_{k_n} \right)$$

$$+\frac{1}{2} \bigotimes_{j=1}^l (I_{k_j} - iI_{k_j}) \bigotimes \left( \frac{1}{2} I_{k_{j+1}} - I_{k_{j+1}} \right) \bigotimes \left( \frac{1}{2} I_{k_n} - I_{k_n} \right). \quad (32)$$

Obviously, the operators $[X_l, (D_0 - D_{N-1})]_\pm$ are $l-$order quantum operators. Therefore, if the integer index $l$ is an even number the operators $[X_l, (D_0 - D_{N-1})]_\pm$ will be even-order multiple-quantum operators. Note
that when the integer index $l$ is an even number the Hermitian operator $X_l = 2^{l-1}I_{k_1x}I_{k_2x}...I_{k_lx}$ also is really an even-order multiple-quantum operator which is a linear combination of the even-order multiple-quantum operators with various quantum orders smaller than $l$ including zero-quantum operators. In a more general case an $l$-order multiple-quantum operator $H_l$ could be further generated from the Hermitian operators $[X_l, (D_0 \pm D_{N-1})]$ and $[X_l, (D_0 \pm D_{N-1})]_\pm$, for example,

$$H_l = \sum_i a_i^\pm [iX_l, (D_0 \pm D_{N-1})] + \sum_i b_i^\pm [X_l, (D_0 \pm D_{N-1})]_\pm.$$ 

To load the information of the marked state $|s\rangle$ on the multiple-quantum operators one may first prepare the operators $\{X_l\}$ or the operator function $f(D_s)$ from the diagonal operator $D_s$ of the marked state $|s\rangle$ and other diagonal operators by Eq.(26) or (27). Then these operators are further used to construct the desired multiple-quantum operators in a similar way to $[X_l, (D_0 - D_{N-1})]_\pm$. Once the desired Hermitian multiple-quantum operator $H_l$ is prepared one can further construct the multiple-quantum unitary operator $U_l(D_s, \lambda_l) = \exp(-i\lambda_l H_l)$. There is a well-known operator identical relation for any two Hermitian operators $A$ and $B$ [33, 34]:

$$\lim_{m \to \infty} (e^{iA/\sqrt{m}} e^{iB/\sqrt{m}} e^{-iA/\sqrt{m}} e^{-iB/\sqrt{m}})^m = e^{-[A,B]}.$$ (33)

This formula and Eq.(30) could be helpful for the decomposition of the multiple-quantum unitary operator into a sequence of the oracle unitary operations and oracle-independent unitary operations. This decomposition could be used as the oracle iterative sequence $U(D_s)$ in the quantum search sequence.

3.3 The Grover iterative sequence in the spin ensemble

According the above scheme of Eq.(24) to construct the oracle unitary sequences $U(D_s)$ a simple oracle unitary sequence $U(D_s)$ is constructed explicitly below. This sequence is similar to the Grover iterative sequence [3, 17]. The time evolution propagator for the oracle unitary sequence (here denote $U(D_s)$ as $U(m)$) is constructed simply by

$$U(m) = [\exp(-i\pi D_{N-1}) \exp(-i\pi D^x_s)]^m$$ (34)

where the oracle-independent unitary operation $\exp(-i\pi D_{N-1})$ is a LOMSO unitary operator and the oracle unitary operation sequence $\exp(-i\pi D^x_s)$ a multiple-quantum unitary operator. The Hermitian multiple-quantum operator $D^x_s$ is defined as
$D^x_s = \exp(-i\frac{\pi}{2}F_y)D_s \exp(i\frac{\pi}{2}F_y) = \bigotimes_{k=1}^{n} \left( \frac{1}{2}E_k + a_k^s I_{kx} \right)$.  

(35)

By using the relation between the two diagonal operators $D_s$ and $D_0$ [17],

$D_s = \exp(-i\frac{\pi}{2}F_x)U_{ox}(-\frac{\pi}{2})D_0 U_{ox}(\frac{\pi}{2}) \exp(i\frac{\pi}{2}F_x)$

(36)

where the auxiliary oracle unitary operation $U_{ox}(\theta) = \prod_{k=1}^{n} \exp[-i\theta a_k^s I_{kx}]$, one can re-express the propagator of Eq.(34) as

$U(m) = \exp(-i\frac{\pi}{2}F_y) \exp(-i\frac{\pi}{2}F_x) \times \left[ \exp(-i\pi D^x_0) \exp(-i\pi D_0) \right]^m U_{ox}(\frac{\pi}{2}) \exp(i\frac{\pi}{2}F_x) \exp(i\frac{\pi}{2}F_y)$.  

(37)

It follows from the definition of the diagonal operator $D_s$ of the marked state that there is an expansion for the unitary operator $\exp(\pm i\pi D_s)$ [17],

$\exp(\pm i\theta D_s) = E + (-1 + \exp(\pm i\theta))D_s$.  

(38)

The iterative part of the propagator $U(m)$ of Eq.(37) then can be simplified by the expansion

$G(m) = \left[ \exp(-i\pi D^x_0) \exp(-i\pi D_0) \right]^m$

$= [E - 2D_0 - 2D_0^x + 4D_0^x D_0]^m$.  

(39)

It can turn out based on the definition of the diagonal operator $D_k$ that the operator set $\{E, D_0, D_0^x, D_0^x D_0, D_0^x D_0^x D_0\}$ form a closed operator algebra subspace. Then the iterative sequence of Eq.(39) can be generally written as

$G(m) = E + \alpha_1(m)D_0 + \alpha_2(m)D_0^x + \alpha_3(m)D_0D_0^x + \alpha_4(m)D_0^x D_0$.  

(40)

Using the relations $G(m + 1) = G(1)G(m) = G(m)G(1)$ one can obtain the recursion equations for the coefficients $\{\alpha_k(m)\}$,

$\begin{pmatrix} \alpha_1(m+1) \\ \alpha_2(m+1) \\ \alpha_3(m+1) \\ \alpha_4(m+1) \end{pmatrix} = \begin{pmatrix} -1 + 4/N & 2/N \\ -2 & -1 \\ -1 & -2/N \\ 2 & -1 + 4/N \end{pmatrix} \begin{pmatrix} \alpha_1(m) \\ \alpha_2(m) \\ \alpha_3(m) \\ \alpha_4(m) \end{pmatrix} + \begin{pmatrix} -2 \\ 0 \\ -2 \\ 4 \end{pmatrix}$  

(41a)

and $\alpha_1(m) = \alpha_2(m)$ and $\alpha_4(m) = -2\alpha_1(m) + \alpha_3(m)$. The recursion equations (41a) and (41b) have the following solution:

$\alpha_1(m) = \alpha_2(m) = -\frac{N}{N-1}[1 - \cos(m\theta)]$,

$\alpha_3(m) = -\frac{N}{N-1}[-1 + \cos(m\theta) + \sqrt{N-1}\sin(m\theta)]$,

$\alpha_4(m) = \frac{N}{N-1}[1 - \cos(m\theta) + \sqrt{N-1}\sin(m\theta)]$  

where $\cos \theta = -1 + 2/N$ and $\sin \theta = 2\sqrt{N-1}/N$.

The time evolution of a spin ensemble under the propagator of Eq.(37) then can be expressed as

$\rho(t) = U(m)\rho(0)U(m)^{-1}$.
The density operator \( \rho \) is traceless. Then the unitary transformation relations:

\[
\begin{align*}
\rho & = \exp(-i \frac{\pi}{2} F_x) \exp(-i \frac{\pi}{2} F_z) U_{ao}(\frac{\pi}{2}) G(m) \\
& \times U_{ao}(\frac{\pi}{2}) \exp(i \frac{\pi}{2} F_y) \rho \exp(-i \frac{\pi}{2} F_y) \exp(-i \frac{\pi}{2} F_x) U_{ao}(\frac{\pi}{2}) \\
& \times G(m)^+ U_{ao}(\frac{\pi}{2}) \exp(i \frac{\pi}{2} F_x) \exp(i \frac{\pi}{2} F_y). \tag{42}
\end{align*}
\]

Now assume that the initial density operator is given by \( \rho(0) = \sum_k \varepsilon_k I_{kz} \).

Then one has the unitary transformation relations:

\[
egin{align*}
U_{ao}(\frac{\pi}{2}) \exp(i \frac{\pi}{2} F_y) \rho(0) \exp(-i \frac{\pi}{2} F_y) \exp(-i \frac{\pi}{2} F_x) U_{ao}(\frac{\pi}{2}) \\
= - \sum_k \varepsilon_k I_{kz},
\end{align*}
\]

\[
G(m) \{ - \sum_k \varepsilon_k I_{kx} \} G(m)^+ = - \sum_k \varepsilon_k \{ I_{kx} + \gamma_1(m) D_0 + \gamma_2(m) D_0^x \\
+ \gamma_3(m) D_0 D_0^x + \gamma_4(m) D_0 D_0^x + \gamma_5(m) D_0 I_{kx} + \gamma_6(m) I_{kz} D_0 \\
+ \gamma_7(m) D_0 D_0 I_{kx} + \gamma_8(m) I_{kz} D_0 D_0 \}
\]

where the coefficients \( \gamma_l(m) \) \((l = 1, 2, \ldots, 8)\) are given by

\[
\begin{align*}
\gamma_1(m) &= \frac{1}{N} [\alpha_1(m)^3 + \alpha_2(m) \alpha_3(m)^* + \alpha_3(m) \alpha_2(m) ], \\
\gamma_2(m) &= \frac{1}{2} [\alpha_2(m) + \alpha_3(m) + \alpha_2(m) \alpha_3(m) ] \\
+ \frac{1}{2} \alpha_2(m)^* \alpha_4(m) + \frac{1}{2} \alpha_2(m) \alpha_4(m)^*, \\
\gamma_3(m) &= \frac{1}{2} \alpha_3(m)^* + \alpha_3(m) \alpha_2(m) + \alpha_2(m) \alpha_3(m) + \frac{1}{2} \alpha_3(m) \alpha_4(m)^* \\
\gamma_4(m) &= \frac{1}{2} \alpha_3(m)^* + \alpha_3(m) \alpha_2(m) + \alpha_2(m) \alpha_3(m) + \frac{1}{2} \alpha_3(m) \alpha_4(m)^*, \\
\gamma_5(m) &= \alpha_1(m), \gamma_6(m) = \alpha_1(m)^*, \\
\gamma_7(m) &= \alpha_4(m), \gamma_8(m) = \alpha_4(m)^*.
\end{align*}
\]

The density operator \( \rho_l(m) \) of Eq.(42) then can be further given by

\[
\begin{align*}
\rho_l(m) &= \sum_{k=1}^n \varepsilon_k \{ I_{kz} - \gamma_1(m) D_0^x - \gamma_2(m) D_{N-1} \\
- \gamma_3(m) D_0 D_{N-1} + \gamma_4(m) D_{N-1} D_0^x + \gamma_5(m) D_0^x I_{kz} \\
+ \gamma_6(m) I_{kz} D_0^x + \gamma_7(m) D_{N-1} D_0^x I_{kz} + \gamma_8(m) I_{kz} D_0^x D_{N-1} \} \tag{43}
\end{align*}
\]

Now the conversion efficiency of the initial density operator \( \rho(0) = \sum_k \varepsilon_k I_{kz} \)
to the multiple-quantum coherence under the Grover iterative sequence of
Eq.(34) can be obtained from Eq.(43). The \( LOMSO \) operator in the density
operator \( \rho_l(m) \) of Eq.(43) which have not any contribution to the multiple-
quantum transitions is given by

\[
\begin{align*}
\rho_z(m) &= \sum_{k=1}^n \varepsilon_k \{ I_{kz} - \frac{1}{N} \gamma_1(m) E - \gamma_2(m) D_{N-1} - \frac{1}{N} \gamma_3(m) D_{N-1} \\
- \frac{1}{N} \gamma_4(m) D_{N-1} + \frac{1}{N} \gamma_5(m) I_{kz} + \frac{1}{N} \gamma_6(m) I_{kz} + \frac{1}{2N} \gamma_7(m) D_{N-1} + \frac{1}{2N} \gamma_8(m) D_{N-1} \}
\end{align*}
\]

The density operator \( \rho_l(m) \) is traceless since the initial density operator \( \rho(0) \) is traceless. Then the \( LOMSO \) operator \( \rho_z(m) \) is reduced to the form
\[ \rho_z(m) = \sum_{k=1}^{n} \varepsilon_k \{ I_{kz}[1 + \frac{1}{N}(\gamma_5(m) + \gamma_6(m))] - \frac{1}{N}\gamma_1(m)E + \gamma_1(m)D_{N-1} \}. \quad (44) \]

In general, the diagonal operator \( D_s \) defined by Eq.(9) can be expanded as a sum of the LOMSO product operator base

\[ D_s = \frac{1}{N} \{ E + \sum_{k=1}^{n} (a^*_k 2I_{kz}) + \sum_{l>k=1}^{n} (a^*_k 2I_{lz})(a^*_l 2I_{lz}) \]

\[ + \sum_{m>l>k=1}^{n} (a^*_k 2I_{kz})(a^*_l 2I_{lz})(a^*_m 2I_{lz}) \]

\[ + \ldots \} \]. \quad (45) \]

Then inserting the expansion (45) of the diagonal operator \( D_{N-1} \) into Eq.(44) one obtains

\[ \rho_z(m) = \sum_{k=1}^{n} \varepsilon_k I_{kz}[1 + \frac{1}{N}(\gamma_5(m) + \gamma_6(m))] \]

\[ + \frac{1}{N} (\sum_{k=1}^{n} \varepsilon_k) \gamma_1(m) \{ \sum_{k=1}^{n} (-2I_{kz}) + \sum_{l>k=1}^{n} (-2I_{kz})(-2I_{lz}) \]

\[ + \sum_{m>l>k=1}^{n} (-2I_{kz})(-2I_{lz})(-2I_{lz}) \]

\[ + \ldots \} \}. \quad (46) \]

Therefore, the conversion coefficient \( C_m \) for the initial density operator (or the initial longitudinal magnetization) \( \rho_I(0) = \sum_{k=1}^{n} \varepsilon_k I_{kz} \) is given by

\[ C_m = 1 + \frac{1}{N}[\gamma_5(m) + \gamma_6(m)] - \frac{2}{N}(\sum_{l=1}^{n} \varepsilon_l/\varepsilon_k)\gamma_1(m). \quad (47) \]

The conversion coefficient \( C_m = 1 \) means that the initial magnetization \( \rho_I(0) \) is not transferred into any other components including multiple-quantum coherence in the spin ensemble, while \( C_m = 0 \) means that the initial magnetization \( \rho_I(0) \) is completely converted into other components in the spin ensemble. It can be seen from the coefficients \( \{\alpha_k(m)\} \) that the maximum \( \gamma_1(m), \gamma_5(m), \gamma_6(m) \sim O(1) \) and occur at \( m \sim \sqrt{N} \). Then the conversion coefficient \( C_m \) of Eq.(47) shows that the initial magnetization \( \rho_I(0) \) can not be transferred effectively into multiple-quantum coherence for a large qubit number \( (N = 2^n) \) and the transferred efficiency \( (1 - C_m) \) decreases exponentially as the qubit number for any iterative number \( m \). This result shows that the Grover iterative sequence (34) is not an effective sequence to enhance the NMR signal and amplify the realizable size of a spin ensemble in solving quantum search problem in a spin ensemble. This is quite different from the pure-state quantum search algorithm which can achieve a quadratic amplification for the amplitude of the marked quantum state. One sees again
the difference between the ensemble quantum search computation and the
pure-state quantum search computation.

3.4 NMR signal enhancement by cross interaction between oracle-
dependent and oracle-independent interactions

The extraction of the solution information of the search problem in the
multiple-quantum spectrum could be simplified by preparing the suitable or-
acle iterative sequence $U(D_s)$ and its effective Hamiltonian $H(D_s)$ used to
excite the multiple-quantum coherence. As an example, assume that the
information of the marked state $|s\rangle$ is loaded on the zero-quantum operator
subspace of the Liouville operator space of the spin system. The preparation
for the zero-quantum Hamiltonian carrying the information of the marked
state can be carried out by using the discrete Fourier analysis and the phase
cycling technique [17], as can be seen in Eq.(28). By starting the proper oper-
ator function $f(D_s)$ of the diagonal operator $D_s$ of the marked state and the
oracle-independent operator function $f(D_r)$ the zero-quantum Hamiltonian
may be constructed generally

$$H_{zq} = \frac{1}{N_1} \sum_{k=0}^{N_1-1} \exp(-i\varphi_k F_z) \left[f(D_s) + f(D_r)\right] \exp(-i\varphi_k F_z)$$

$$= H_{zq}(D_s) + H_{zq}(D_r) \quad (48)$$

and the corresponding zero-quantum unitary operator is already given by
Eq.(29). If the oracle-independent Hamiltonian $H_{zq}(D_r)$ is suitably cho-
sen, for example, it may be chosen as those zero-quantum coherence of the
$k$–qubit $(k \leq n)$ subsystem of the spin system and moreover, as the dominat-
ing term in Eq.(48) with respect to the Hamiltonian $H_{zq}(D_s)$, then extraction
of the solution information could be simplified and the zero-quantum coherence
which carry the information could be enhanced in amplitude, as can
be seen below. Now there are three types of the zero-quantum coherence
in the spin ensemble. One type is of the $k$–qubit subsystem of the spin
system, another of the rest $(n - k)$–qubit subsystem, and the third is those
cross zero-quantum coherence between the two subsystem. The last type
is more important. Assume that all spins of the $k$–qubit subsystem have
the same precession frequency and so do all spins of the rest $(n - k)$–qubit
subsystem, but the two precession frequencies are different each other. Then
the cross zero-quantum coherence between the two subsystems may have
precession frequencies different from zero frequency that is the precession
frequency of the zero-quantum coherence of each subsystem. Therefore, the
cross zero-quantum coherence with nonzero precession frequencies and those
zero-quantum coherence of the two subsystems can be discriminated in the
zero-quantum spectrum. Since these cross zero-quantum peaks also contain
the information of the marked state one could obtain the information of the
marked state from the spectral parameters of the cross zero-quantum peaks.
The zero-quantum peaks of the \( k \)-qubit subsystem are strongest since the
oracle-independent Hamiltonian \( H_{zq}(D_r) \) is the dominating term in the
total Hamiltonian \( H_{zq} \), while those peaks of the \((n - k)\)-qubit subsystem are
usually weakest and even cannot be observable if a small call number of the
oracle unitary operation is used in the sequence, but the information of the
marked state may not be obtained from these zero-quantum peaks because
of overlapping of the strong zero-quantum peaks without the information.
However, amplitude for the cross zero-quantum coherence is dependent on
both the oracle-dependent and oracle-independent Hamiltonians. Then the
cross zero-quantum peaks could be much stronger than those zero-quantum
peaks of the \((n - k)\)-qubit subsystem and could be observable even when the
peaks of the \((n - k)\)-qubit subsystem are too weak to be observable. Therefore,
one may use these cross zero-quantum peaks with nonzero precession
frequencies to determine the solution of the search problem. Obviously, this
NMR signal enhancement is achieved by the cross interaction between the
oracle-dependent and oracle-independent Hamiltonians. To see more clearly
the enhancement mechanism one first expresses the zero-quantum unitary
transformation in the interaction frame as

\[
U_{zq}(t) = \exp(-iH_{zq}t) = \exp(-itH_{zq}(D_r))T \exp(-i \int_0^t H'_{zq}(D_s, t')dt')
\]  

(49)

where \( T \) is Dyson time-ordering operator and the oracle-dependent Hamiltonian in the interaction frame is given by

\[
H'_{zq}(D_s, t) = \exp[itH_{zq}(D_r)]H_{zq}(D_s) \exp[-itH_{zq}(D_r)].
\]  

(50)

Equation (49) may be used to build up the quantum circuit \( U_{zq}(t) \) which
equals \( U(D_s) \) in the quantum search sequence [37]. The interaction Hamiltonian (50) is responsible for generating the cross zero-quantum peaks. It can be expanded formally as

\[
H'_{zq}(D_s, t) = H_{zq}(D_s) + it[H_{zq}(D_r), H_{zq}(D_s)]
- \frac{1}{2}t^2[H_{zq}(D_r), [H_{zq}(D_r), H_{zq}(D_s)]] + \ldots.
\]  

(51)

One sees that there are the cross interaction terms between the two Hamiltonians \( H_{zq}(D_r) \) and \( H_{zq}(D_s) \) in the higher-order terms of the expansion except the term \( H_{zq}(D_s) \). These cross interaction terms are responsible for generating the cross zero-quantum peaks. Obviously, the cross interaction in strength is dependent on the oracle independent Hamiltonian \( H_{zq}(D_r) \), and a
strong Hamiltonian $H_{zq}(D_r)$ will generate a strong cross interaction. Therefore, a strong oracle-independent Hamiltonian would enhance greatly the cross zero-quantum peaks since strong cross interaction would create strong cross zero-quantum peaks. On the other hand, the oracle-independent Hamiltonian $H_{zq}(D_r)$ becomes more and more important in the higher order terms, as can be seen in Eq.(51). A cross interaction term with a high order such as $[H_{zq}(D_r), ..., [H_{zq}(D_r), H_{zq}(D_s)]]$ could amplify the effect of the oracle-independent Hamiltonian on the cross zero-quantum peaks.

Generally, the Baker-Campbell-Hausdorff (BCH) formula [38] can be exploited to construct the higher-order cross interaction. For convenience, operators $A = H(D_r)$ and $B = H(D_s)$ denote the oracle-independent and oracle-dependent Hamiltonians, respectively. The operator $A$ may be any Hermitian operator of the spin system and usually is taken conveniently as the LOMSO operator. The oracle-dependent Hamiltonian $H(D_s)$ is usually taken as an Hermitian multiple-quantum operator. The BCH formula [35, 36, 38] shows that the symmetric composition for the two operators $A$ and $B$ with the second order approximation is written as

$$S_A(x) = \exp(xA') \exp(xB) \exp(xA') = \exp(xA_1)$$

with the Hermitian operator,

$$xA_1 = x(A + B) + \frac{1}{6}x^3([B, [B, A']] - [A', [A', B]]) + ...,$$

where $x = it$ is the imaging time and $A' = A/2$ (the same denoting for operator $B'$, $A'_1$, ..., below). There is also another second order symmetric composition with a different product order of the operators $A$ and $B$,

$$S_B(x) = \exp(xB') \exp(xA) \exp(xB') = \exp(xB_1)$$

with the Hermitian operator,

$$xB_1 = x(A + B) + \frac{1}{6}x^3([A, [A, B']] - [B', [B', A]]) + ....$$

Because the unitary operators $S_q(x) (q = A, B)$ of Eqs. (52a) and (53a) satisfy the time symmetry: $S_q(x)S_q(-x) = E$ (E is unity operator) there are not even-order commutators such as $[A, B]$, $[A, [A, B]]$, etc., in the composed operators $A_1$ and $B_1$ of Eqs. (52b) and (53b) [36]. Here an $n$-order cross interaction term is defined as a $(n + 1)$-order commutator of the operators $A$ and $B$. Then the second-order cross interaction, which is defined as the third order commutators such as $[A, [A, B]]$ and $[B, [B, A]]$, can be built up in the same symmetric form as Eq.(52a) with the operators $A_1$ and $B_1$ again according to the BCH formula,

$$\exp(xA'_1) \exp(-xB_1) \exp(xA'_1) = \exp(xA_3)$$

where the cross interaction $A_3$ is given by

$$xA_3 = x(A_1 - B_1) + \frac{1}{6}x^3([-B_1, [-B_1, A'_1]] - [A'_1, [A'_1, -B_1]]) + ...$$
\[ = 1/8 x^3([B, [B, A]] - [A, [A, B]]) + ..., \quad (54b) \]
and
\[ \exp(xB_1') \exp(-xA_1) \exp(xB_1') = \exp(xB_3) \quad (55a) \]
where the cross interaction \( B_3 \) is given by
\[ xB_3 = x(-A_1 + B_1) + 1/6 x^3([-A_1, [-A_1, B_1]] - [B_1', [B_1', -A_1]]) + ... \]
\[ = 1/8 x^3([A, [A, B]] - [B, [B, A]]) + ... \quad (55b) \]
It can be seen in Eqs. (54b) and (55b) that the operators \( A \) and \( B \) disappear and the dominating terms become the third order commutators \([A, [A, B]]\) and \([B, [B, A]]\) in the cross interaction \( A_3 \) and \( B_3 \). If those commutators with order higher than four can be neglected in Eqs. (54b) and (55b) then the operators \( A_3 \) and \( B_3 \) are approximated as the second order cross interaction. The fourth-order cross interaction can be prepared by starting the second order cross interaction \( A_3 \) and \( B_3 \),
\[ \exp(xA_3') \exp(xB_3) \exp(xA_3') = \exp(xA_5) \quad (56a) \]
and
\[ \exp(xB_3') \exp(xA_3) \exp(xB_3') = \exp(xB_5) \quad (56b) \]
The higher order cross interaction also can be easily built up with the same method above. The complexity of the quantum circuit to construct the higher order cross interaction is usually dependent on the two Hamiltonians \( H(D_r) \) and \( H(D_s) \). However, it is easy to see that preparing \( 2m \)-order cross interaction terms \( A_{2m+1} \) and \( B_{2m+1} \) need \( 1/2(3m+1 - 1) \) and \( 1/2(3m+1 + 1) \) oracle unitary sequences \( \exp(xB) \), respectively. How high order cross interaction \( A_{2m+1} \) and \( B_{2m+1} \) is required for an \( n \)-qubit spin system so that the initial density operator \( \rho(0) = \sum_k \epsilon_k I_{kz} \) can be transferred efficiently into the multi-qubit quantum coherence by the oracle unitary sequence \( U(D_s) = \exp(-iA_{2m+1}) \exp(-iB_{2m+1}) \)? This is also dependent on the two Hamiltonians \( H(D_r) \) and \( H(D_s) \) but it could be lower than \( n \) for the proper \( H(D_r) \) and \( H(D_s) \). A much more efficient method to prepare the higher order cross interaction can be seen in [35, 36]. According to the method [35, 36] one first composes in a symmetric form the \((2m - 1)\)-order composed unitary operators \( f_{2m-1}^A(x) \) and \( f_{2m-1}^B(x) \) by using the second order composed unitary operators \( S_A(x) \) and \( S_B(x) \) of Eq. (52a) and (53a), respectively,
\[ f_{2m-1}^A(x) = \exp[x(A + B) + x^{2m+1} A_{2m+1} + ...] = S_A(p_1 x)S_A(p_2 x)...S_A(p_r x), \quad (57a) \]
\[ f_{2m-1}^B(x) = \exp[x(A + B) + x^{2m+1} B_{2m+1} + ...] = S_B(p_1 x)S_B(p_2 x)...S_B(p_r x) \quad (57b) \]
where the parameters \( \{ p_r \} \) must be determined suitably and satisfy \( \sum_{k=1}^{r} p_k = 1 \), \( p_j = p_{r+1-j} \) (\( j = 1, 2, \ldots, r-1 \)), and it is usually not easy to determine the parameters \( \{ p_r \} \) for a higher-order composition [35, 36]. In Eqs.(57a) and (57b) it needs about \( r \approx 2^m \) unitary operators \( S_A(x) \) (\( S_B(x) \)) to compose the unitary operator \( f_{2m-1}^A(x) \) (\( f_{2m-1}^B(x) \)) for small \( m \) [35]. Note that there is difference of the product order of the operators \( A \) and \( B \) in the operators \( f_{2m-1}^A(x) \) and \( f_{2m-1}^B(x) \). The difference is necessary to construct further the \( 2m \)-order cross interaction with the operators \( f_{2m-1}^A(x) \) and \( f_{2m-1}^B(x) \). Again one uses the above symmetric BCH formula to compose further the \( 2m \)-order cross interaction:

\[
\exp(\frac{1}{2}x(A + B) + \frac{1}{2}x^{2m+1}B_{2m+1} + ...) \exp(-x(A + B) - x^{2m+1}A_{2m+1} + ...) \\
\times \exp(\frac{1}{2}x(A + B) + \frac{1}{2}x^{2m+1}B_{2m+1} + ...) = \exp[-x^{2m+1}(A_{2m+1} - B_{2m+1}) + ...].
\]

(58)

Obviously, here the parameter \( x \) does not take a small value so that the \( 2m \)-order cross interaction \( A_{2m+1} \) or \( B_{2m+1} \) could be large enough to derive the conversion of the initial density operator to the multiple-quantum coherence in a spin ensemble. This is different from the conventional decomposition of exponential operators [33, 35, 36] where the parameter \( x \) is always taken as a small quantity.

Here leave two questions of the quantum search NMR multiple-quantum pulse sequence in the future. One is how high order cross interaction is necessary to excite effectively the multiple-quantum coherence in an \( n \)-qubit spin ensemble. Another is how to extract experimentally the information of the marked state from the multiple-quantum spectrum excited by the higher order cross interaction.

4. Discussion

The quantum mechanical unitary dynamical description for the Turing machines was first given by Benioff [39] in 1980. This quantum Turing machine model has not proved to be superior to the classical counterparts, as pointed out by Deutsch [2]. However, the quantum mechanical unitary dynamics plays an important role in computational speedup in ensemble quantum computation in contrast to in the pure-state quantum computation. A direct result of the quantum mechanical unitary dynamics is that
the quantum search problem and the parity problem could be solved in polynomial time on an NMR ensemble quantum computer, while these problems are NP problems and can not be solved efficiently in classical computation. Therefore these works really move the frontier between the solvability and intractability of complex problems. There is difference between ensemble quantum computation based on the quantum mechanical unitary dynamics and the pure state version. For example, in the paper it has been shown that the Grover algorithm is an ineffective iterative sequence used to amplify the output signal of the NMR ensemble quantum computation, while it can achieve a quadratic amplification for the amplitude of the marked state in the pure-state quantum search. The reason for this is that the iterative sequence of the Grover algorithm can amplify quadratically the relative amplitude of the marked state and suppress any other states in the quantum system, but the detectable NMR signal intensity in a spin ensemble is not only proportional to the relative amplitude of the marked state but also the ensemble-averaging magnetization strength of the spin system. The ensemble quantum computation based on the quantum mechanical unitary dynamics allows the separation each other between the initial input state and the quantum circuit in a quantum algorithm. This makes it possible for the highly mixed states of the spin ensemble to be used in ensemble quantum computation. This also shows that there is difference between new ensemble quantum computation and the classical NMR quantum computation working on the concept of the pseudopure state or the effective pure state.

In the paper it has been shown that the quantum search problem could be solved in polynomial time on an NMR quantum computer. On the basis of the quantum mechanical unitary dynamics the NMR multiple-quantum spectroscopy has been developed to solve experimentally the quantum search problem. In the quantum search sequence based on the NMR multiple-quantum spectroscopy the information of the marked state is first loaded on the unitary evolution propagator that excites the multiple-quantum coherence from the initial density operator, i.e., a highly mixed state, of the spin ensemble. This propagator is usually constructed with the oracle unitary operation and oracle-independent unitary operations. The NMR multiple-quantum spectra that carry the information of the marked state then are obtained experimentally. Finally one could extract the information of the marked state from the multiple-quantum spectra by the conventional NMR data processing. Although number of the multiple-quantum coherence or transitions increases exponentially as the qubit number in a spin ensemble,
number of the multiple-quantum spectral peaks can be controlled and can be as low as \((2n + 1)\) in the multiple-quantum spectra for an \(n\)-qubit spin system. This ensures that at least some lower-order multiple-quantum peaks do not reduce exponentially as the qubit number as long as the initial density operator is transferred effectively to the multiple-quantum coherence by the oracle iterative sequence of the quantum search sequence. The complexity of the NMR multiple-quantum experiments is therefore mainly dependent on the oracle iterative sequence exciting the multiple-quantum coherence and the extraction of the information of the marked state from the multiple-quantum spectra. The NMR multiple-quantum spectroscopy could provide a more convenient and larger space to design new ensemble quantum computation sequences or experiments. It also could provide possible approaches to enhancing the output NMR signal of ensemble quantum computation and extending the realizable size of the NMR ensemble quantum computation. In particular, in the frame of the multiple-quantum spectroscopy the cross interaction between oracle-dependent and oracle-independent interactions has been proposed to enhance the output NMR signal of the ensemble quantum computation. In comparison with those NMR quantum search algorithms or experiments [11, 17, 40] the quantum search sequence based on the quantum mechanical unitary dynamics and the NMR multiple-quantum spectroscopy would be more powerful.

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