Quantum information processing using strongly-dipolar coupled nuclear spins

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An important issue in experimental quantum information processing (QIP) is achieving coherent control while increasing the number of qubits \cite{1, 2}. In all existing implementations of quantum computation, the execution time is limited by durations of nonlocal gates. In the case of nuclear magnetic resonance (NMR) implementations of QIP, where qubits are formed by mutually coupled spin 1/2 nuclei, most of the existing implementations used isotropic liquids. In these systems, the durations of nonlocal gates are limited by the strength of (indirect) scalar couplings, which typically are of the order of 10-100 Hz.

Apart from the scalar couplings, nuclear spins also interact through magnetic dipole-dipole couplings, which are 2-3 orders of magnitude stronger than the scalar couplings, and would therefore allow significantly faster gate operations. In the case of isotropic liquids, the rapid molecular reorientation averages the dipolar interactions to zero. On the other hand, in oriented systems, the dipolar interactions survive and are therefore potentially strong enough to be used instead of direct) scalar couplings, which are typically of the order of 10-100 Hz.

The Hamiltonian for a dipolar coupled n-spin system is,

\[
H = \sum_{j=1}^{n} \hbar \omega_j I_j^z + \sum_{j=1,k<j}^{n} 2\pi \hbar D_{jk} (3I_j^z \cdot I_k^z - I_j^z \cdot I_k^z)
\]  

(1)

where \(\omega_j\) are the chemical shifts, \(D_{jk}\) are the dipolar coupling constants and \(I^z_j\) are \(z\)-components of the spin angular momentum operators \(I_j^z\). The first term corresponds to the Zeeman interaction and the second term describes the dipolar interaction. When \(|D_{jk}| \ll |\omega_j - \omega_k|\), one usually invokes the ‘weak-coupling’ approximation \cite{3, 4}, where one ignores the off-diagonal parts of the Hamiltonian. Then the Zeeman product states are the eigenstates of the full Hamiltonian and individual spins can be conveniently treated as qubits. Most of the experiments in NMR-QIP have so far been carried out on such systems \cite{2}.

However, to optimize the execution speed of our quantum processor it is desirable to use stronger couplings, including \(|D_{jk}| \geq |\omega_j - \omega_k|\). In this case, we have to use the complete Hamiltonian (1), where the Zeeman and the coupling parts do not commute and not all eigenstates are Zeeman product states but linear combinations of them. Unlike in the case of a weakly coupled spin system, individual spins of a strongly coupled system loose addressability. In such cases the eigenstates of the full Hamiltonian can form individually controllable subsystems and then the eigenbasis becomes the natural and accurate choice for the computational basis.

Though strongly dipolar coupled spin-systems have been suggested for QIP earlier \cite{3}, so far it had not been possible to implement general unitary gate operations that can be applied to arbitrary initial conditions. Here we show how quantum computation can be realized on the eigenbasis by efficient coherent control techniques. As a specific system, we use a strongly dipolar coupled four-spin system partially oriented in a liquid crystal. Such systems have certain key merits. Unlike in the liquid state systems, the intramolecular dipolar couplings are not averaged out completely, but are only scaled down by the order parameter of the solute molecules oriented in the liquid crystal \cite{5, 6}. Since intermolecular interactions are averaged out (unlike the crystalline systems), liquid crystalline systems provide well defined quantum registers with low decoherence rates. We achieve high-fidelity coherent control with the help of specially designed strongly modulating pulses (SMPs).

To demonstrate these techniques, we use an interesting algorithm suggested by S. P. Jordan \cite{9}: the quantum algorithm for numerical gradient estimation (QNGE). Before we discuss our implementation, we summarize the theoretical description of QNGE \cite{9}. The gradient of an one-dimensional real function \(f\) over a small real range \(l\) is written as \(\nabla f = \{f(l/2) - f(-l/2)\}/l\). Thus classically two function evaluations are necessary to estimate the gradient of a one-dimensional function and a minimum of \(d + 1\) function evaluations are necessary for a \(d\)-dimensional function. On the other hand, QNGE requires only one function evaluation independent of the dimension of the function. Here the function \(f\) is encoded in an \(n\)-qubit input register. An ancilla register is also required whose size \((n_0 \text{ qubits})\) depends on the maximum possible value of the gradient.

In QIP, numbers are represented in the form of a register. To encode the real number \(x\) in the input register, we have to convert it into a nonnegative integer \(\delta \in \{0, 1, \cdots , N-1\}\),
where $N = 2^n$. The encoding is defined by

$$x = \frac{l}{N-1} \left( \delta - \frac{N-1}{2} \right).$$

(2)

The circuit diagram for the quantum algorithm is shown in Figure 1. Initially the ancilla register ($an$) is set to 1 and the input register ($in$) is set to 0. The inverse quantum Fourier transform (IQFT) prepares a plane wave state on the ancilla register and the Hadamard transform (H) prepares a uniform superposition on the input register $|10\rangle$:

$$|00\cdots00\rangle \xrightarrow{\text{IQFT-an}} H^{in} \xrightarrow{\text{H}} |01\cdots00\rangle.$$

The ancilla register is now in an eigenstate of the addition modulo $N_0$. On applying oracle $U_f : |an, in\rangle \rightarrow |sf(x) \oplus N_0, an, in\rangle$, where $s$ is a scaling factor, the total state becomes

$$\frac{1}{\sqrt{N_0 N}} \sum_{k=0}^{N_0-1} e^{-i\frac{2\pi k}{N_0}} |k\rangle \sum_{\delta=0}^{N-1} e^{i\frac{2\pi \delta s f(x)}{N}} |\delta\rangle.$$

(3)

For a small $x$, $f(x) \approx f(0) + x \nabla f$. Substituting for $x$ from equation 2 and ignoring the global phase, the input register reduces to,

$$\frac{1}{\sqrt{N}} \sum_{\delta=0}^{N-1} e^{i\frac{2\pi \delta \cdot \nabla f}{N}} |\delta\rangle.$$

The scaling factor $s$ is set to maximize the precision i.e.,

$$s = N_0(N - 1)/Nl.$$

(3)

Now a phase estimation is carried out with the quantum Fourier transform (QFT) on the input register

$$\frac{1}{\sqrt{N}} \sum_{\delta=0}^{N-1} e^{i\frac{2\pi \delta \cdot \nabla f}{N}} |\delta\rangle \xrightarrow{\text{QFT}} \nabla f.$$

Measuring now the input registers in the computational basis gives an estimation of $\nabla f$.

In our 4-qubit case, we select two qubits as ancillas and the other two as input qubits i.e., $n_0 = n = 2$, $N_0 = N = 4$, and $\delta \in \{00, 01, 10, 11\}$. From equation (2), $x \in \{-1/2, -1/6, 1/6, 1/2\}$ for $l = 1$. Let us consider an example function $f(x) \in \{0, 2/3, 4/3, 2\}$, which has a gradient $\nabla f = 2$. Using equation 3 we obtain the scaling factor $s = 3$ so that $sf(x) \in \{0, 2, 4, 6\}$. For $\delta \in \{00, 10\}$, $sf(x) \oplus N_0$ remains identity. For $\delta \in \{01, 11\}$, $sf(x) \oplus N_0$, $sf(x) \oplus N_0$ adds 2 to the ancilla, i.e., flips the first ancilla qubit. Therefore, for this example, the oracle is a CNOT($an_{i1}, in_{i2}$) gate i.e., a NOT operation on the first ancilla qubit controlled by the second input qubit. The propagator for the entire algorithm is therefore,

$$U_{QNGE} = U_{\text{QFT}}^{in} \cdot U_{\text{CNOT}}(an_{i1}, in_{i2}) \cdot U_{H}^{in} \cdot U_{\text{IQFT}}^{in}.$$

We design a single SMP corresponding to this entire operation on the input states.

An SMP is a cascade of radio-frequency (RF) pulses numerically calculated based on the precise knowledge of the internal Hamiltonian of the qubit system and the target propagator $U_T$. Fortunato et al [11] have described numerically searching for a target propagator $U_T$. Given a target operator $U_T$, Fortunato et al [11] have described numerically searching for an SMP propagator $U_{\text{SMP}}$ based on four RF parameters for each segment $k$: duration ($\tau^{(k)}$), amplitude ($\omega^{(k)}_A$), phase ($\phi^{(k)}$) and frequency ($\omega^{(k)}_F$). We found that it is useful to add one more degree of freedom: after each pulse segment, we introduce a variable delay $\tau^{(k)}_d$. The delays are computationally inexpensive to optimize, easy and accurate to implement and make designing non-local gates easier. All the parameters are determined so as to maximize the fidelity

$$F = \text{trace} \left[ U_T^{-1} \cdot U_{\text{SMP}} \right] / M.$$
specific to a known initial state. The fidelity of an SMP is determined from the experimental and the calculated spectra are less than 0.1 Hz and 6% respectively. The elements of the diagonalized Hamiltonian are shown in Figure 3. The line width range from 1.7 Hz to 4.0 Hz indicating coherence times ($T_2^*$ relaxation times) between 1.8 s and 0.7 s. The coherence times are sufficiently long to ignore relaxation effects in the design of the SMP.

The procedure for analyzing the NMR spectra of partially oriented systems has been well studied. We have developed a numerical procedure to iteratively determine the system Hamiltonian from its spectrum and a guess Hamiltonian. The 37 strongest transitions of the CIB spectrum were used and a unique fit was obtained. The mean frequency and intensity errors between the experimental and the calculated spectra are less than 0.1 Hz and 6% respectively. The elements of the diagonalized system Hamiltonian and the corresponding energy level diagram are shown in Figure 4.

In NMR-QIP, the initial states are not pure states but are pseudopure states that are isomorphic to pure states. The pseudopure states differ from the pure states by a uniform background population on all states. It is easier, however, to prepare a pair of pseudopure states (POPS) instead of a single qubit gate. We prepared the pair $|0100⟩⟩0100⟩ − |0000⟩⟩0000⟩$. The first term represents the desired initial state; the additional second part does not interfere with the QNGE experiment, because the operation IQFT, when acted on $|0⟩$, creates a uniform superposition of the ancilla qubits. Such a state is invariant under the $f(x)⊕N_0$ operation and therefore the output state corresponding to $|0000⟩⟩0000⟩$ is independent of the oracle $U_f$. The inset

$F' = \frac{\text{trace}[\rho_T \cdot \rho_{\text{SMP}}]}{\sqrt{\text{trace}[\rho_T^2] \cdot \text{trace}[\rho_{\text{SMP}}^2]}}$
In the state at the end of the quantum algorithm, the input qubits are encoded into three sets each of 15 linearly independent states. The results of the diagonal tomography obtained, under the mean of the diagonalization condition, there are 15 unknowns for the 16 eigenvalues. The populations are then measured using a linear PFG and average over several (32) transients. Therefore we use a random delay (in between 0 and 10 ms) after the PFG and average over several (32) transients. The populations are then measured using a linear detection (small flip-angle) pulse. Using the normalization condition, there are 15 unknowns for the 16 eigenstates. The results of the diagonal tomography obtained by the mean of three sets each of 15 linearly independent transitions are shown in Figure 4.

After the projective measurement in the eigenbasis at the end of the quantum algorithm, the input qubits are in the state $|10\rangle$ encoding the gradient $\nabla f = 2$, while the ancilla qubits have equal probability in all possible states. Therefore, the theoretical output diagonal state of the combined system is $I_{an} \otimes (|10\rangle\langle10| - |00\rangle\langle00|)$, where $I_{an}$ is the identity operator for the ancilla qubits and the two parts in the parenthesis correspond to the two parts of the POPS. The diagonal correlation $C$ between the theoretical density matrix $(\rho_T)$ and the experimental density matrix $(\rho_E)$, is defined as

$$C = \frac{\text{trace}[|\rho_T| \cdot |\rho_E|]}{\sqrt{\text{trace}[|\rho_T|^2] \cdot \text{trace}[|\rho_E|^2]}}$$

where $|$ denotes extraction of the diagonal part. The average diagonal correlations were 0.999 and 0.979 for the POPS and the result of the QNGE, respectively. The lower value of the latter may be attributed to decoherence and spectrometer nonlinearities.

In conclusion, we have demonstrated quantum computation on the eigenbasis of system Hamiltonian using coherent control techniques. An example, we described the first implementation of Jordan’s algorithm for numerical gradient estimation. Compared to the usual approach using weakly coupled systems, the present method using strongly dipolar coupled systems yields significantly faster execution times and is therefore less susceptible to decoherence effects. From molecular and spectroscopic considerations a combination of homo and heteronuclear spins in either liquid crystalline or molecular single crystalline environments is a natural way to build larger qubit-systems (with $> 10$ qubits). The coherent control of strongly dipolar coupled systems then becomes important and the present work is the first step in this direction. We believe that the coherent control techniques demonstrated here for dipolar coupled nuclear spins will turn out to be essential also for other solid-state implementations of quantum information processing.

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