Non-thermal nuclear magnetic resonance quantum computing using hyperpolarized xenon

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Current experiments in liquid-state nuclear magnetic resonance (NMR) quantum computing are limited by low initial polarization. To address this problem, we have investigated the use of optical pumping techniques to enhance the polarization of a 2-qubit NMR quantum computer ($^{13}$C and $^1$H in $^{13}$CHCl$_3$). We have generalized the procedure for effective pure state preparation in order to efficiently use the increased polarization. With this more flexible scheme, an effective pure state is prepared with polarization enhancement of a factor of 10 compared to the thermal state. An implementation of Grover’s quantum search algorithm with a polarization-enhanced spin system is demonstrated. © 2001 American Institute of Physics. [DOI: 10.1063/1.1409279]

Intensive experimental efforts have been made to implement quantum computing since Shor and Grover developed their respective algorithms. So far, the only experimental demonstrations of quantum computing algorithms have been based on nuclear magnetic resonance. However, the low polarization of high-temperature nuclear spin systems is a major limitation of this approach. To cope with this situation, contributions of highly mixed spin state, schemes have been developed which produce an effective pure state. Unfortunately, all of these techniques require exponential resources to implement, and improved scalable procedures for fully polarizing an embedded spin system are not practical with the very low initial polarizations found in nuclear magnetic resonance quantum computation (NMRQC). In addition to the experimental challenges, recent theoretical work has called into question the boundary between classical and quantum computing in the case of highly mixed systems. A crucial parameter in determining where this boundary lies is the initial polarization of the quantum system. It is therefore highly relevant to explore existing techniques used to enhance polarization for NMR and to investigate their application to quantum computing.

One recently developed technique is based on laser-polarized xenon. It is known that the high electron polarization in optically pumped alkali metals like rubidium can be transferred to the nuclear spins of noble gases such as $^3$He or $^{129}$Xe via a spin-exchange mechanism. More recently, it was found that hyperpolarized liquid $^{129}$Xe could be used to enhance the polarization of nuclei of other molecules in solution via the spin polarization-induced nuclear Overhauser effect (SPINOE). The focus of our work is the coupling of optical pumping with NMRQC. We have developed a temporal labeling scheme which is more flexible than the existing ones and which allows efficient use of the polarization enhancements. To validate this technique, we have created an effective pure state with an order of magnitude polarization enhancement. The flexibility of our scheme is further illustrated by applying this approach to perform Grover’s search algorithm. Polarization enhancements up to a factor of 7 are achieved for the final computed state.

In a typical optical pumping experiment, we start by pressurizing a glass cell (Pyrex, 200 cc) containing solid Rb to 3.5 atm with a mixture of ultrapure gases: 12% natural abundance Xe, 2% $^2$N, and 86% He. After the cell is heated to 110°C to produce a saturated Rb vapor, a 100 W diode array laser (Optopower), tuned to the D1 transition of Rb, is switched on. This circularly polarized light irradiates the gas mixture and polarizes the Rb electrons. After 20 min of Rb–Xe spin exchange, the gas mixture is passed through a cold trap which condenses the highly polarized Xe. The Xe is then melted and refrozen in a high-pressure NMR tube (New Era) containing 20 μl of degassed $^{13}$CHCl$_3$. The sample tube is moved into a 2.1 T magnet and the temperature regulated to $-40°C$ so that $^{13}$CHCl$_3$ and Xe liquefy and mix (the Xe:$^{13}$CHCl$_3$ molar ratio = 5:1 to 8:1). The hyperpolarized liquid $^{129}$Xe (typical polarization of 1%–2%) enhances the polarization of $^1$H and $^{13}$C in $^{13}$CHCl$_3$ via the SPINOE effect. Results are monitored using a homebuilt spectrometer (see Fig. 1). We have achieved polarizations over a factor of 10 greater than the thermal equilibrium value for both $^1$H and $^{13}$C.

The deviation density matrix $\rho^{\text{dev}} = \rho - aI$, with $\text{tr}(\rho^{\text{dev}})$...
schemes cyclically permute (\( P_i \)) the \( {1H}^{13}C \) spin state at time \( t_1 \). The elements on the diagonal \(((00), ..., (11))\) are experimental data \(((0) (1))\) corresponding to the ground state, and therefore Eq. 1 allows one to derive the variables \( w_i \).

We applied this generalized temporal labeling procedure to an \( n = 2\)-qubit quantum computer with polarization-enhanced initial states. The density matrix \( \rho_{\text{init}} \) is inferred by adding a probing experiment a short time before the permutation experiment. The probing experiment consists of two simultaneous rf pulses with small tip angle \((10^\circ - 20^\circ)\) on both spins and leads to results like those shown in the inset of Fig. 1. The time \( r_1 \) is calculated to be long enough so that the system has returned to its quasi-equilibrium enhanced state, yet short enough \((r_1 = 25 s \approx T_{1,Xe} = 15 s)\) so that the initial density matrix inferred from the probing experiment closely approximates the state \( \rho_{\text{init}} \), at the start of the permutation experiment. The full effective pure state preparation thus consists of the preparation of three \((2^n - 1)\) optically pumped samples. On each sample, a permutation experiment is performed, preceded by a probing experiment to gain information on \( \rho_{\text{init}} \) such that the weights \( w_i \) in Eq. (1) can be calculated. The weighted sum of the results of the permutation experiments then produces \( \rho_{\text{eff}} \). The polarization enhancement of \( \rho_{\text{eff}} \) over the effective pure state produced at thermal equilibrium can be determined (see Fig. 2). An effective pure state with a factor of 9.5 polarization enhancement was created.

We further illustrate the flexibility of our generalized temporal labeling scheme by performing multiple experiments using one optically pumped sample. In terms of polarization, this can be done since Xe, which has a very long relaxation time \((T_{1,Xe} = 15 min)\), is a quasi-continuous source of high polarization. After each experiment, which lasts less than 1 s, the time for \( {13}CHCl_3 \) to return to a quasi-equilibrium enhanced state is 2 min \((5T_{1,CHCl_3})\). Therefore, multiple experiments can be performed before the xenon polarization relaxes significantly. Thus, instead of preparing three separate optically pumped samples, only one sample is prepared on which three permutation experiments are performed (see typical starting times \( t_1, t_2, \) and \( t_3 \) in Fig. 1). The \( \rho_{\text{init}} \) now significantly different since the polarization is decreasing and hence a set of weights is definitely needed to create an effective pure state. It can be proven that, as long as the set of \( \rho_{\text{init}} \) are diagonal matrices, Eq. (1) represents the temporal labeling procedure with optimal signal to noise (proof similar to that shown in Ref. 5). This variation of the labeling scheme was used to create the input states of a quantum computation.

We demonstrated this technique by implementing Grover’s search algorithm; unlike the Deutsch–Josza algorithm, \( \rho_{\text{init}} \) this requires a pure input state to produce a meaningful result.

\[
\begin{align*}
\rho_{\text{eff}} &= \sum_{i=0}^{2^n-1} w_i (P_i \rho_{\text{init},i} P_i^\dagger),
\end{align*}
\]

Provided one can determine \( \rho_{\text{init},i} \) experimentally, the optimal set of permutations \( P_i \) can be chosen and the weights can be calculated from Eq. (1). The latter is done by considering that \( \text{diag}(\rho_{\text{eff}}) \) represents a vector with constant elements \( b \) except at the position corresponding to the ground state and therefore Eq. (1) yields a set of \( 2^n - 1 \) linear equations in the \( 2^n - 1 \) variables \( w_i/b \). Comparison of the signal to noise with that of in the thermal case (all \( w_i = 1 \)) allows one to derive the variables \( w_i \).

FIG. 1. Deviation density matrix (the diagonal is shifted to obtain a zero average for the nonground states) of the effective pure state.
and therefore is more demanding. The goal of the 2-qubit Grover algorithm is to identify an element \( x_0 \) among four possible elements \( x_i \) by querying an oracle function \( f(x) \) for which \( f(x_0) = 1 \) while \( f(x_i \neq x_0) = 0 \). The four elements \( x_i \) are represented by the spin states \( |00\rangle, |01\rangle, |10\rangle, |11\rangle \) of \( ^1\text{H}13\text{C} \). Classically this search would take an average of 2.25 attempts, while one query is sufficient using the Grover algorithm.\(^{14,15}\) The output of Grover’s search algorithm is the state \( |x_0\rangle \). The experimental procedure is similar to effective pure state preparation, except that the permutation experiment is now replaced by a concatenation of the pulses of the cyclic permutation \( P_1 \) and the pulse sequence representing the actual quantum algorithm (the same protocol as used in Ref. 14). The resulting \( ^1\text{H} \) and \( ^{13}\text{C} \) readout spectra are compared with the thermal spectra (Fig. 3). We successfully implemented the four possible cases \( x_0 = 00, 01, 10, \) or 11 of Grover’s quantum algorithm with polarization enhancements of the final computed states as large as a factor of 7.

In conclusion, we have demonstrated that, with optical pumping and using our generalized temporal labeling scheme, a polarization-enhanced effective pure state can be produced and a quantum computation can be performed. The polarization enhancements of more than a factor of 10 for both \( ^{13}\text{C} \) and \( ^1\text{H} \) in \( ^{13}\text{CHCl}_3 \) are comparable to other published results using hyperpolarized \( ^{129}\text{Xe} \) and SPINOE.\(^{11}\) Even though large scale NMR quantum computers are not yet within reach, the first steps for the necessary polarization enhancement have now been taken. Further increases in polarization can be achieved by using isotopically pure \( ^{129}\text{Xe} \), which increases the polarization by a factor of 3–4, by improvements in the design of the pumping apparatus, as well as by the screening of other candidate quantum computing molecules. Moreover, effective control over a large range of initial polarizations could allow one to use NMRQC to explore the fundamental divergence between quantum computing and classical computing.

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