Non-thermal nuclear magnetic resonance quantum computing using hyperpolarized Xenon
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Current experiments in liquid-state nuclear magnetic resonance 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. In this paper, we present a new approach to improve the polarization of the system by applying this approach to perform Grover’s search algorithm. Polarization-enhancements up to a factor of 7 are achieved.

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% CHCl, 2% N₂ 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 frozen (77K) in a high-pressure NMR tube (New Era) containing 20μl of degassed 13CHCl₃. The sample tube is moved into a 2.1T magnet and the temperature regulated to −40°C so that 13CHCl₃ and Xe liquefy and mix (Xe:13CHCl₃ molar ratio = 5:1 to 8:1). The hyperpolarized liquid ¹₂⁹Xe enhances the polarization of ¹H and ¹³C in ¹³CHCl₃ via the SPINOE effect. Results are monitored using a home-built spectrometer (see Fig. 1). The data show that the enhancements are large in the Xe-¹³CHCl₃ mixture. We have achieved polarizations over a factor of 10 greater than the thermal equilibrium value for both ¹H and ¹³C.

The deviation density matrix \( \rho = qI + \rho_{\text{dev}} \) of the \( (¹H+¹³C) \) spin state at time \( t_1 \) is shown in the inset to Fig. 1. The data were obtained by observing ¹H and ¹³C simultaneously after sending two small tip angle pulses, providing full information about the diagonal elements of the deviation density matrix \( \rho_{\text{dev}} \). Even though the polarizations of ¹³C and ¹H are larger than in the thermal case, the spin state is still highly mixed.

Labeling schemes are applied to create an effective pure state from a mixed state \( \rho_{\text{eff pure}} = q_1 I + q_2 \rho_{\text{pure}} \). Temporal labeling schemes cyclicly permute \( (P) \) the populations of all states but the ground state. The resulting \( 2^n - 1 \) density matrices – with \( n \) the number of quantum bits – are summed to create an effective pure state \( \rho_{\text{eff pure}} = \sum_{i=1}^{2^n-1} P \rho_{\text{pure}} P^i \). If the initial state is reproducible and its density matrix diagonal in the computational basis, the optimal choice of ground state (in
of the 2 enhancement from experiment to experiment and therefore coherent). However, there are small changes in initial relaxation, the mechanism underlying SPINOE, is not enhanced initial states are known to be diagonal (cross-relaxation, the mechanism underlying SPINOE, is not coherent). However, there are small changes in initial enhancement from experiment to experiment and therefore the initial density matrices are not all identical for each of the $2^n - 1$ cyclic permutation experiments. Therefore, we generalized the existing temporal labeling scheme by introducing weights $w_i$ in the summation and allowing $\rho_{\text{init}}$ to vary from experiment to experiment:

$$\rho_{\text{eff pure}} = \sum_{i=1}^{2^n-1} w_i (P_i \rho_{\text{init},i} P_i^\dagger).$$

Provided one can determine $\rho_{\text{init},i}$ experimentally, the weights can be calculated since the matrix representation of Eq. 1 yields a set of $2^n - 1$ linear equations in the $2^n - 1$ 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 $\rho_{\text{init},i}$ is inferred by adding a probing experiment a short time $t_1$ before the permutation experiment. The probing experiment consists of two simultaneous RF pulses with small tip angle ($10^\circ$ to $20^\circ$) on both spins leading to results like those shown in the inset to Fig. 1. The time $t_1$ is calculated to be long enough so that the system has returned to its quasi-equilibrium enhanced state, yet short enough ($r_1 = 25 \text{ sec} \ll T_{1,\text{Xe}} = 15 \text{ min}$) so that the density matrix inferred from the probing experiment closely approximates the state $\rho_{\text{init},i}$ 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 $\rho_{\text{init},i}$. Then Eq. 1 is solved for $w_i$ and the enhancement of $\rho_{\text{eff pure}}$ over the effective pure state resulting from experiments at thermal equilibrium is 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 ($T_{1,\text{Xe}} = 15 \text{ min}$), is a quasi-continuous source of high polarization. After each experiment, which lasts less than a second, the time for $\text{CHCl}_3$ to return to a quasi-equilibrium enhanced state is 2 min ($5T_{1,C,H}$). Therefore, multiple experiments can be performed before the Xenon polarization relaxes significantly. Thus, instead of preparing 3 separate optically pumped samples, only one sample is prepared on which 3 permutation experiments are performed (see typical starting times $t_1$, $t_2$ and $t_3$ in Fig. 1). The $\rho_{\text{init},i}$ are now significantly different since the polarization is decreasing, but a set of weights $w_i$ can still be calculated from Eq. 1 to create an effective pure state. It can even be proven that as long as the set of $\rho_{\text{init},i}$ are diagonal matrices, Eq. 1 represents the temporal labeling procedure with optimal signal-to-noise ratio. This variant 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, this requires a pure input state to produce a meaningful result 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 query-
FIG. 3. Spectral readout of the four cases of the 2-qubit Grover search using the polarization-enhanced initial state of $^{13}$CHCl$_3$. The plots show the real part of the $^1$H (left) and $^{13}$C (right) spectra, with NMR lines at $\pm \frac{1}{2} J_{CH}$ (in Hz relative to $\nu_H$ and $\nu_C$). Positive (negative) peaks correspond to the state $|0\rangle$ ($|1\rangle$). The vertical scale is arbitrary. From top to bottom the marked element is $x_o = 00, 01, 10$ or 11 and as the spectra show the Grover algorithm outputs exactly this element. The data are obtained by using our generalized temporal labeling scheme and by performing multiple experiments with one optically pumped sample. Each instance of the Grover algorithm is executed twice, reading out first the $^1$H and then the $^{13}$C spectra. The numbers on the plots represent the polarization-enhancements achieved. They vary between 2 and 7.

Using an oracle function $f(x)$ for which $f(x_o) = 1$ while $f(x_i \neq x_o) = 0$. The four elements $x_i$ are represented by the spin states $|00\rangle, |01\rangle, |10\rangle, |11\rangle$ of $\{^{1}$H$^{13}$C$\}$ ($|0\rangle$ and $|1\rangle$ correspond to nuclear spin state up and down respectively). Classically this search would take an average of 2.25 attempts, while one query is sufficient using the Grover algorithm $\begin{bmatrix} 4 & 4 \\ 1 & 1 \end{bmatrix}$. The output of Grover’s search algorithm is the state $|x_o\rangle$. Using our generalized labeling scheme, the experimental implementation starts with a probing experiment to gain information about $p_{\text{init}}$. A time $\tau_1$ later the quantum computation experiment is performed, which is a concatenation of the pulses of the cyclic permutation $P_i$ and the pulse sequence representing the actual quantum algorithm (same protocol used in $\begin{bmatrix} 4 & 4 \\ 1 & 1 \end{bmatrix}$). The resulting $^1$H and $^{13}$C readout spectra are compared with the thermal spectra (Fig. 3). We successfully implemented the four possible cases $x_o = 00, 01, 10$ or 11 of Grover’s quantum algorithm with polarization enhancements 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 $^{13}$C and $^1$H in $^{13}$CHCl$_3$ are comparable to other published results using hyperpolarized Xe and SPINOE $\begin{bmatrix} 4 & 4 \\ 1 & 1 \end{bmatrix}$. 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 $^{129}$Xe, which increases the polarization by a factor of 3 to 4, by improvements in the design of the pumping apparatus, as well as by the screening of other candidate quantum computing molecules. Moreover, the effective control over a large range of initial polarizations could allow NMRQC to explore the fundamental divergence between quantum computing and classical computing.

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