Quantum information processing using localized ensembles of nuclear spins

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We describe a technique for quantum information processing based on localized ensembles of nuclear spins. A qubit is identified as the presence or absence of a collective excitation of a microscopic ensemble of nuclear spins surrounding a single quantum dot. All single and two-qubit operations can be effected using hyperfine interactions and single-electron spin rotations, hence the proposed scheme avoids gate errors arising from entanglement between spin and orbital degrees of freedom. Ultra-long coherence times of nuclear spins suggest that this scheme could be particularly well suited for applications where long lived memory is essential.

Nuclear spin degrees of freedom have attracted considerable attention as potential carriers of quantum information due to their exceptionally long coherence times. Early bulk NMR work\textsuperscript{[1]} has substantially enriched our understanding of the key features of quantum computation\textsuperscript{[2, 3, 4, 5, 6]}. The fundamental difficulties in scaling bulk NMR to a large number of qubits motivated efforts to use single, individually addressable nuclear spins in semiconductors as qubits\textsuperscript{[4]}. Two-qubit operations in Ref.\textsuperscript{[2]} rely upon exchange coupling, making them susceptible to fast orbital decoherence mechanisms.

Recently, a method for robust storage of quantum information in localized ensembles of nuclear spins was suggested\textsuperscript{[6, 10]}, where it was shown that the collective hyperfine coupling between nuclear and electron spin degrees of freedom provides a controllable mechanism for coherent storage and manipulation of quantum states. These nuclear spin ensembles correspond, for example, to the lattice nuclei in a quantum dot. As a quantum memory, such nuclear ensembles are robust with respect to variations in dot characteristics, rely upon proven fabrication techniques, and provide high fidelity storage without requiring a high-degree of nuclear spin polarization.

In this Letter, we describe a technique to efficiently process quantum information stored in localized nuclear spin ensembles. Specifically, these ensembles enable a robust, scalable implementation of quantum computation protocols, unencumbered by the difficulties faced by single spin impurity or bulk NMR approaches. The fundamental interaction that allows for spin manipulation in our scheme is hyperfine coupling: as a result, the orbital and spin degrees of freedom remain unentangled throughout the two-qubit gate operations, mitigating the effects of orbital decoherence on gate fidelity. While collective enhancement of hyperfine interaction allows for fast quantum gates, the ultra-long nuclear spin coherence times render the scheme particularly attractive for memory intensive quantum information processing tasks.

The scheme proposed here can be realized using either electrically\textsuperscript{[8]} or optically\textsuperscript{[9]} manipulated quantum dots that are defined as in Fig.\textsuperscript{1} The nuclear ensemble in each dot is prepared using polarized electron spins\textsuperscript{[6, 10]}. We illustrate our technique by considering the fully polarized case (when the nuclear state is $|0⟩ = |-I, \ldots, -I⟩$ for $N$ spin-$I$ nuclei), demonstrate how to perform single qubit and two-qubit gates, and consider sources of error.

The Hamiltonian describing a single electron interacting with the nuclear spins in a quantum dot is\textsuperscript{[11]}

\begin{equation}
H = \hbar B \gamma_S \hat{S}_z + \hbar B \sum_k \gamma_l^{(k)} I_z^{(k)} + \hbar a \hat{S} \cdot \hat{A}.
\end{equation}

The first and second terms describe the coupling of the
electron and the nuclear spins to an external magnetic field, with effective gyromagnetic ratios $\gamma_S, \gamma_I^{(k)}$. The last term is the electron coupling to collective nuclear degrees of freedom, defined by $\hat{A}_{\pm z} = \sum_k a_k \hat{I}_{z}^{(k)}$. The $a_k$'s are proportional to the probability density of the electron at the location of the corresponding nuclei and are normalized such that $\sum_k a_k = N$; $a = A/N$ is the average hyperfine interaction per nucleus. Both $A$ and $N$ depend upon the specific material and dot construction; typical numbers are $A \sim 10^{-100}$ ns$^{-1}$ and $N = 10^4 - 10^6$ nuclei. Excitations out of the fully polarized state form an orthonormal set of collective nuclear spin states $|m\rangle \propto (\hat{A}_z)^m |0\rangle$, where $m$ is the number of excitations.

When an electron is confined in the dot, evolution over times much shorter than $a^{-1}$ is restricted to subspaces spanned by $\{|m-1\rangle \uparrow, |m\rangle \downarrow\}$. Using Pauli matrices, the Hamiltonian for each subspace with a fixed excitation number $m$ is $H^{(m)} = \hbar \delta m \sigma_z^{(m)} + \hbar \Omega m \omega_x^{(m)}$, with

$$\Omega_m = a/2 \sqrt{(m-1) \hat{A}_- \hat{A}_+ |m-1\rangle};$$

$$\delta_m = a/4 (|m\rangle \hat{A}_z |m\rangle + |m-1\rangle \hat{A}_+ |m-1\rangle) + |\gamma_S - (|m\rangle K_z |m\rangle - |m-1\rangle K_z |m-1\rangle|B/2,$$

where $K_z = \sum \gamma_I I_z^{(k)}$ is a sum over nuclear spin operators, weighted by individual nuclear spin gyromagnetic ratios. When Overhauser shift and Zeeman energy sum to zero ($|\delta_m| \ll \Omega_m$) and coherent flip-flop (Rabi) oscillations occur at rate $\Omega_m$. The energy level structure of the coupled electron-nuclear system is shown in Fig. 2 along with the coupling strengths for $m \leq 2$: since $\Omega_{m+1} = \eta_m \Omega_m$ where $\eta_m = \sqrt{m+1} - \mathcal{O}(m/N)$, it is easy to note the analogy with the celebrated Jaynes-Cummings (JC) model of quantum optics [12]. We use the nonlinearity of such a JC-type two-level system coupled to a nearly-bosonic mode to effect elementary quantum gates.

Quantum information stored in the $m = 0, 1$ manifold can be mapped reliably from nuclear states to electron spin and back [8] via a generalized rotation:

$$R_{e,n}^{(\theta, \phi)}(\pi/2, 0)(\alpha|0\rangle + \beta|1\rangle) |\downarrow\rangle = |0\rangle (\alpha |\downarrow\rangle + i\beta |\uparrow\rangle),$$

where

$$R_{e,n}^{(\theta, \phi)} = e^{i\phi \hat{S}_z} e^{-i\theta \hat{H} / (\hbar \Omega_e)} e^{-i\phi \hat{S}_z}.$$  

The transfer of quantum information from the nuclear ensemble to electron spin allows for fast single qubit operations to be performed: after a $|\downarrow\rangle$ electron is injected into the dot, the quantum information is transferred to the electron spin, and then the operation is performed on the electron. Finally, the quantum information is mapped back to the nuclear ensemble. A $z$-axis rotation can be accomplished by waiting in a static magnetic field or through a laser-induced spin-dependent AC Stark shift [10]. $x$-axis rotations can be done via

$$\begin{align*}
&|2, \downarrow\rangle &\rightarrow &\eta \Omega_1 &|1, \uparrow\rangle \\
&|1, \downarrow\rangle &\rightarrow &\Omega_{ESR} &|0, \uparrow\rangle \\
&|0, \downarrow\rangle &\rightarrow &\Omega_{ESR} &|1, \uparrow\rangle
\end{align*}$$

FIG. 2: Level structure of the combined electron spin and nuclear dark state plus excited states. For $N \rightarrow \infty$, the coupling between the two excitation manifold is stronger by $\eta \rightarrow \sqrt{2}$.

ESR [8, 13] (with $\Omega_{ESR} \gtrsim 1$ ns$^{-1}$) or optical Raman spin flips through a virtual trion state (with $\Omega_{opt, ESR} \gtrsim 10$ ns$^{-1}$) [8]. Measurement of the ensemble nuclear spin state can be implemented by mapping the quantum information to electron spin, and carrying out an electron spin measurement either by state selective ionization followed by charge measurement with a rf-SET [14] or by detecting fluorescence in a spin-dependent cycling transition [13, 10].

To perform a two-qubit gate between quantum dots A and A', a single electron can be used as to transfer quantum information between the dots: the state of nuclear spin qubit A is mapped onto the electron, which is then moved to A', where a controlled-phase (CP) gate between the nuclear and the electronic qubit is applied using the nonlinearity of the interaction. Following Ref. [17], a two-qubit CP-gate (up to single qubit gates) is given by

$$R_{e,n}^{(\pi/4, 0)} R_{e,n}^{(\pi/\eta, -\pi/2)} R_{e,n}^{(-\pi/4, 0)}.$$  

In the computational basis, this corresponds to

$$\begin{pmatrix}
1 & 0 & 0 & 0 \\
0 & e^{i\pi/\eta} & 0 & 0 \\
0 & 0 & e^{-i\pi/\eta} & 0 \\
0 & 0 & 0 & -1
\end{pmatrix}.$$  

After this operation, the quantum information carried by the electron spin is mapped back onto the nuclear spin A. Applying a $z$-rotation of $-\pi/\eta$ to qubit A and one of $\pi/\eta$ to qubit A' yields a CP gate.

For distant dots, where electron spin transport between ensembles is difficult or impossible, it may be easier to generate entanglement off-line, apply local purification protocols and then use it to effect non-local gates, following [13]. Entanglement between distant electron spins can be generated optically, using spin-flip optical Raman transitions [13], or electronically, such as through adiabatic splitting of a spin singlet in a double dot [20]. The electron-nuclear state mapping procedure can then ensure that the distant nuclear spin ensembles are entangled. Starting with the entangled nuclear state $|\downarrow\rangle (|01\rangle + |10\rangle)$ (between dots A' and B', cf. Fig. 1), we can implement a CP gate on dots A and B (deterministically) by performing local unitaries and measurements.
of AA’ and BB’ as follows: (i) perform a CNOT\(_{A \rightarrow A’}\) then measure A’. (ii) Perform a CP gate \(|10\rangle \rightarrow -|10\rangle\) at BB’ followed by a Hadamard gate and measurement at B’. (iii) Local phase flips \(|0\rangle \rightarrow -|0\rangle\) at A (B) if the measurement outcomes at B’(A’) were “1” complete the CP gate between A and B [21].

We next analyze various sources of errors due to finite polarization, inhomogeneity, nuclear spin dynamics, and electron spin decoherence. To understand the role of finite polarization, the specific cooling procedure must be examined. When the nuclear ensemble starts as a thermal mixture, cooling to dark states can be achieved by coupling polarized electron spins to the nuclear ensemble [10]. Regardless of the details of cooling, the final density matrix will be a statistical mixture of dark states \(|\mathcal{D}(n, \beta)\rangle\), where \(A \cdot |\mathcal{D}(n, \beta)\rangle = 0\). \(n\) is the number of spin excitations (\(n = 0\) is the fully polarized state) and \(\beta\) is the permutation group quantum number [22]. It was recently shown [3] that dark states have the same symmetry properties as the fully polarized state, and a manifold of excited states can be defined from a given dark state, \(|m(n, \beta)\rangle \propto (\hat{A}_z)^m |\mathcal{D}(n, \beta)\rangle\). Hence the above considerations for perfectly polarized nuclei map directly to the case when the nuclear ensembles start in any given dark state, not just the fully polarized state.

In practice, the cooled nuclear ensemble density matrix is a mixture of different dark states, i.e., \(\hat{\rho} = \sum_{n, \beta} \rho_{n, \beta} |\mathcal{D}(n, \beta)\rangle \langle \mathcal{D}(n, \beta)|\). As each dark state has a different \(\delta_m(n, \beta), \Omega_m(n, \beta)\), interaction times and applied magnetic fields can only produce \(R^{xy}_{en}(\theta, \phi)\) with the desired angle \(\theta\) for some fraction of the given mixture. The inhomogeneous mixture effects can be characterized by examining the subgroups of dark states with different detunings, which lead to errors in Rabi oscillations \(p \simeq (\sigma_\delta/\Omega_\epsilon)^2\), where \(\sigma_\delta\) is the standard deviation of possible \(\delta_1\) values over the distribution \(\rho_{n, \beta}\). In the homogeneous case with spin-1/2 nuclei,

\[
\sigma_\delta \simeq a \sqrt{N(1 - P^2)}. \tag{7}
\]

Even at high \((P \sim 0.95)\) polarizations, the effect of different detunings can be substantial \((p \sim 0.03)\). This provides the strongest limit to realization of \(R^{xy}_{en}\) [22].

The inhomogeneous nature of the hyperfine coupling leads to further errors. In this case, the logical states of the system (the m-excitation manifolds) are no longer eigenstates of \(\hat{J}_z = \hat{A}_z + \hat{S}_z\) and of \(\hat{A}^2\). As a consequence, there is a nonzero probability that the system moves out of the computational subspace during the gate operation. We estimate these leakage errors using the techniques developed in Refs. [3, 6], and find that the resulting gate error \(p_{\text{inhom}}\) decreases with increasing number of nuclei: for \(N \sim 10^5\) at high polarizations \((P > 0.95)\) \(p_{\text{inhom}} \lesssim 10^{-3}\) [24]. We note that a similar error emerges due to the differences in Zeeman energy associated to different nuclear species. For materials like GaAs, with gyromagnetic ratios varying greatly from species to species, this limits the effectiveness of gate operations at high magnetic field, resulting in the errors in the range of \(10^{-3}\) as indicated in Fig.3. Optical manipulation (e.g., tuning the system into resonance via spin-dependent AC optical Stark shifts) may mitigate this difficulty. Finally, in between gate operations the errors associated with inhomogeneous evolution may be eliminated by refocusing sequences of NMR pulses.

The nuclear spin diffusion due to dipole-dipole interactions with rate \(\gamma_{DD} \sim 60\text{ms}^{-1}\) leads to a decay of the coherences which form the dark states [22]. Active corrections can be performed with NMR pulse sequences that average the dipole-dipole Hamiltonian, such as WaHuHa, improving the error rate to \(4\text{~ns}^{-1}\) [24, 27]. After correction dark state coherences could have lifetimes on the order of 0.1-1s for moderate cycle times \(\tau_{whh}\). Quadrupolar terms due to inhomogeneous strain can cause additional differential phase evolution of different nuclear spins. However, this type of phase inhomogeneity leads to an error second order in the interaction which is negligible \((\sim 10^{-7}\) per cycle of computation).

The errors of manipulation of single electron spins in quantum dots via microwave or optical fields have been considered in detail elsewhere [3, 4, 13, 17], and only the relevant results are quoted here. Electron spin decoherence will most likely be limited by different Overhauser shifts corresponding to different detunings, with error going as \(p \sim (\sigma_\delta/\Omega_\text{ESR})^2\); fast ESR \((\sim 6\text{~ns}^{-1})\) will mitigate this effect; with optical fields, even faster effective Rabi-oscillations are predicted \((\Omega_\text{opt,ESR} \sim 50\text{~ns}^{-1})\) with errors then limited by spontaneous emission to \(\sim 10^{-3}\). As for measurement, the fundamental limit will be set by relaxation of the electron spin, which has a time scale \(\Gamma^{-1} \sim 0.1-10\text{~ms}\), based on recent measurements of the spin relaxation time [14, 24].

Moving individual electron spins over short distances incurs phase errors due to uncertainty in the local nuclear field of the transport channel. The error induced by randomly oriented nuclei during transit can be estimated by the time-averaged hyperfine field the electron wavepacket encounters over the whole process, \(A/\sqrt{nL\hbar l}\), where \(n\) is the density of lattice nuclei, \(L\) is the length of the channel, and \(\hbar l\) is the transverse area of the channel. For a 50nm wide, 10nm high channel, and a quantum dot separation of 1nm, the expected dephasing probability due to thermal nuclei is \(\simeq 5 \times 10^{-5}\) for a 3ns transfer time.

We consider three materials with demonstrated electronic and optical quantum dots (GaAs, \(N = 10^5\); InAs, \(N = 10^4, 6\); CdSe, \(N = 10^4\)). The expected error of a two-qubit gate operation, defined as \(\text{Tr}[U_{\text{perfect}}U_{\text{actual}}^\dagger]/4\), where \(U_{\text{perfect}}\) is given by Eqn. 6 is plotted in Fig.8. As the error is dominated by detuning error, it is material independent, and is a few percent at 95% polarizations.

To combat these errors, a series of measurements may be made to determine the effective detuning of the system better than the thermal mixture limit \(\sigma_\delta\). This can
The distribution is then narrowed, with \( \tilde{\sigma}_3 \simeq \sigma_3/\sqrt{n} \), where \( n \) is the number of measurements. An improvement in the uncertainty of the mixture’s detuning of a factor of 10 yields high fidelity operation, as shown in the inset to Fig. 3. Smaller dots produce better results at higher polarizations due to their greater coupling strengths. The limits for GaAs are due to the large species inhomogeneity and the incommensurate requirement of sufficient magnetic field to perform effective coherent averaging of dipole-dipole interactions. Low species inhomogeneity allows for higher magnetic field, faster WaHuHa-type correction sequences, and fewer errors in the transfer operation. This would be the case for quantum dots defined in nanotubes with isotopically enhanced \( ^{13}\text{C} \) or in CdSe quantum dots. Materials with low spin-orbit interaction will also reduce electron spin dephasing.

In conclusion, we have detailed a scheme for quantum information processing using dynamically defined qubits composed of collective excitations of nuclear spins in a quantum dot. For small dots with near-homogeneous Zeeman splittings (e.g. CdSe, InAs) large but finite polarization (95%) is already sufficient to reach an error rate in two-qubit operations of order a few percent. Given the suppression of dipolar diffusion in lithographically isolated structures (e.g. vertical quantum dots, nanowhiskers, self-assembled quantum dots) these polarizations may be within reach. Moderate polarizations have already been achieved, and techniques to improve upon this have been considered. Our calculations indicate that the dominant source of error may be mitigated through narrowing the mixture of dark states.

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