Deterministic preparation of Dicke states of donor nuclear spins in silicon by cooperative pumping

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For donor nuclear spins in silicon, we show how to deterministically prepare various symmetric and asymmetric Dicke states which span a complete basis of the many-body Hilbert space. The state preparation is realized by cooperative pumping of nuclear spins by coupled donor electrons, and the required controls are in situ to the prototype Kane proposal for quantum computation. This scheme only requires a sub-gigahertz donor exchange coupling which can be readily achieved without atomically precise donor placement, hence it offers a practical way to prepare multipartite entanglement of spins in silicon with current technology. All desired Dicke states appear as the steady state under various pumping scenarios and therefore the preparation is robust and does not require accurate temporal controls. Numerical simulations with realistic parameters show that Dicke states of 10 – 20 qubits can be prepared with high fidelity in presence of decoherence and unwanted dynamics.

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I. INTRODUCTION

Because of the ultra-long quantum coherence time, electron and nuclear spins of shallow donors in silicon are ideal candidates as information carrier in quantum computation. For $^{31}$P donor in isotope purified $^{28}$Si, a nuclear spin coherence time exceeding 1 s and an electron spin coherence time exceeding 10 ms were measured.\textsuperscript{[3,4]} In his seminal paper, Kane proposed an architecture for scalable quantum computation using nuclear spins of gated $^{31}$P donors in silicon as qubits. With the superb compatibility with the existing silicon technology, Kane’s proposal and its variants have stimulated extensive interests in donor systems in silicon,\textsuperscript{[5–8]} and remarkable technological progresses have since been made in various aspects. Accurate positioning of the $^{31}$P donor was made possible by controlled single-ion implantation.\textsuperscript{[9]} Local electric tuning of the hyperfine interaction between donor electron and nuclear spins was demonstrated.\textsuperscript{[10,11]} An architecture that integrates single $^{31}$P donor with a silicon SET was recently developed which enables high-sensitivity readout and control of the donor electron spin.\textsuperscript{[12–14]} In the meantime, challenges still remain, e.g. on realizing efficient donor electron exchange coupling which is also needed for mediating pairwise interaction of nuclear spins.\textsuperscript{[15]} Due to the interference between the degenerate valleys of the electron, the exchange coupling strength vastly oscillates with sub-nanometer variation in donor position (e.g. between 1-100 GHz when donor separation is $\sim$ 10 nm)\textsuperscript{[4]} The chance to have an exchange coupling of $\geq$ 30 GHz expected by the initial Kane proposal becomes random unless with atomically precise dopant placement\textsuperscript{[15]} which is beyond the technological capability today and in the near future.

In this paper, we introduce a scheme to deterministically prepare Dicke states of donor nuclear spins in silicon. Dicke states in general refer to the common eigenstates of $J^2$ and $J_z$ with eigenvalues $J(J+1)$ and $M$ respectively, where $J$ is the collective spin of an ensemble of $N$ spin-I particles. Dicke states with $J$ taking the maximum value $N/2$ are symmetric under permutation operations. When $M \neq \pm J$, symmetric Dicke states are a class of genuine multipartite entangled states which have been widely pursued in atomic and optical systems as important resources for quantum information processing.\textsuperscript{[20–23]} These states have the remarkable properties that the entanglement is robust against qubit loss and projective measurements on the states lead to various entangled states of a lower qubit number.\textsuperscript{[23]} When $J$ takes value other than $N/2$, the Dicke states are no longer symmetric under all permutation operations and will be referred here as asymmetric. Asymmetric Dicke states are also resources of genuine multipartite entanglement which are less studied for the lack of preparation schemes. The ideal resource for optimal quantum teleportation algorithm is one such state with $J = 0$\textsuperscript{[24]} Preparation of asymmetric Dicke states also makes it possible to access the decoherence free subsystems in the presence of collective decoherence.\textsuperscript{[25]} The scheme we propose here can deterministically access all Dicke states (symmetric and asymmetric) that span a complete basis for the Hilbert space of $N$ spins. This direct access to the Dicke-states basis can be an important complement to the circuit model quantum information processing, since the entanglement of these collective states can not be achieved in a simple way by pairwise interaction\textsuperscript{[26]}.

Our scheme is based on cooperative pumping of nuclear spins by the coupled donor electrons, and the required controls are in situ to the prototype Kane proposal: (i) initialization of electron spin to its ground state in magnetic field; (ii) ac electrical control of the hyperfine coupling of the donor (A-gate); (iii) on and off switching of exchange coupling between neighboring electrons (J-gate). Remarkably, our scheme only needs a sub-gigahertz exchange coupling, which can be satisfied for almost all donor pairs with separation $\sim$ 10 nm. Hence it provides a practical way for generating the critical resource of multipartite entanglement of spins in silicon, which can tolerate the exchange oscillation problem and be realized within the current technology. Our scheme is a significant example of the conceptually new approach of dissipative quantum state preparation with the advantages of robustness and no need for accurate temporal controls as compared to conventional state preparation by coherent evolution, as the desired Dicke states all appear as the unique steady state under the various pumping scenarios. Numerical simulation with realistic parameters for $^{31}$P donors shows that Dicke states of
10 − 20 qubits can be prepared with high fidelity in the presence of decoherence and unwanted dynamics. The scheme also applies to other donor systems with larger nuclear spins such as $^{209}$Bi in silicon\cite{27,29}.

II. CONTROL SCHEME

Fig. 1(a) schematically illustrates the Kane architecture where shallow donors are embedded under patterned electrodes. The A-gate tunes the hyperfine interaction $H_{hf} = \sum_n a_n \hat{\sigma}_n \cdot \hat{I}_n$ with $\hat{\sigma}_n$ and $\hat{I}_n$ being respectively the electron spin and nuclear spin of the $n$th donor. The coupling strength $a_n$ is proportional to the electron density at the donor nucleus site and hence is a function of the voltage applied to the A-gate which pulls the electron wavefunction away from the nucleus. With each A-gate independently controlled with voltage $V_n = V_{n,0} + \delta V_n(t) \cos(\omega t + \phi_n)$, the hyperfine interaction becomes:

$$\hat{H}_{hf} = \sum_n a_n \hat{\sigma}_n \cdot \hat{I}_n + \frac{\partial a}{\partial V} \delta V_n \cos(\omega t + \phi_n) \hat{\sigma}_n \cdot \hat{I}_n. \quad (1)$$

Where the first term is the static hyperfine coupling where $a_{n,0}/2\pi \equiv a(V_{n,0})/2\pi \simeq 60$ MHz, and the second term is the ac hyperfine coupling from the voltage modulation. In a strong magnetic field, the off-diagonal part of the static hyperfine is far off resonance, hence it only results in small shifts of energy levels which are negligible here. However, the off-diagonal part of the ac hyperfine can efficiently pump nuclear spin polarization when the modulation frequency $\omega$ is resonant to the electron-nuclear flip-flop transition\cite{10,12}.

The exchange coupling between neighboring donor electrons is tuned by the J-gate which can separate the electrons. When the electrons are decoupled, nuclear spins of different donors can be independently pumped to the fully polarized state (i.e. nuclear spin initialization). When the electrons are coupled, nuclear spins can be cooperatively flipped by collective raising/lowering operators. We have previously proposed using such pumping to probabilistic prepare singlets of nuclear spins\cite{22,23}.

In general, for coupled donor electrons in uniform magnetic field along $z$ direction, one can always find a non-degenerate one-spin-excitation eigenstate $|\epsilon_0\rangle \equiv \sum_n a_n \hat{\sigma}_n^\dagger |g\rangle$, with $|g\rangle \equiv |\downarrow \cdots \downarrow\rangle$ denoting the ground state. With the ac control frequency $\omega$ tuned near resonance with the transition between $|g\rangle$ and $|\epsilon_0\rangle$, we can neglect all other far detuned electron spin resonances. Dropping non-secular terms, the full Hamiltonian of the electron nuclear spin system can then be written as $\hat{H} = \hat{H}_0 + \hat{H}_c$ where:

$$\begin{align*}
\hat{H}_0 &= \omega_\epsilon |\epsilon_0\rangle \langle \epsilon_0 | + \omega_N \sum_n \hat{I}_n^z, \\
\hat{H}_c &= e^{i\epsilon(t)}|\epsilon_0\rangle \langle \epsilon_0 | \sum_n \Omega_n(t) e^{i\phi_n} \hat{I}_n^z + h.c. .
\end{align*} \quad (2)$$

$\omega_\epsilon$ is the electron resonance frequency between $|\epsilon_0\rangle$ and $|g\rangle$ in the magnetic field, $\omega_N$ is the nuclear Zeeman frequency, and $\Omega_n \equiv \frac{\partial a}{\partial V} \delta V_n$. The dc voltage of each A gate is set such that $a_{n,0} = a_0$. $\hat{H}_c$ induces electron-nuclear flip-flop where the nuclear spin flip is in a cooperative form determined by the phases $\phi_n$ and amplitudes $\delta V_n$ of the ac controls. For simplicity, we consider hereafter a uniformly coupled Heisenberg ring with the eigenstate $|\epsilon_0\rangle = \sum_n \frac{1}{\sqrt{N}} (-1)^n \hat{\sigma}_n^z |g\rangle$ which is gapped from other one-spin-excitation states by $\Delta$ (Fig. 1(b)). Then we have $\Omega_n(t) = (-1)^n \Omega(t)$ where $\Omega = \sqrt{\frac{\partial a}{\partial V}} \delta V$.

We consider two scenarios for the phases $\phi_n$. In the first scenario, $(-1)^n e^{i\phi_n} = 1$ for all donors and hence the electron-nuclear flip-flop term becomes: $\hat{H}_c = \Omega(t) |\epsilon_0\rangle \langle \epsilon_0 | \hat{J}_z + h.c.$ Here $\hat{J}_z = \hat{J}_x \pm i \hat{J}_y$ where $\hat{J} \equiv \sum_n \hat{I}_n$ is the collective spin of all nuclei. In the second scenario, $(-1)^n e^{i\phi_n} = -1$ for a set of donors (referred as group A) while $(-1)^n e^{i\phi_n} = 1$ for the rest (referred as group B), and hence the electron-nuclear flip-flop term is of the form: $\hat{H}_c = \Omega(t) |\epsilon_0\rangle \langle \epsilon_0 | (\hat{J}_A - \hat{J}_B) + h.c.$, where $\hat{J}_A$ and $\hat{J}_B$ are the collective spin of nuclei.
in group A and group B respectively, and \( \hat{J}_A + \hat{J}_B = \hat{J} \). Dicke states here refer to the common eigenstates of \( \hat{J}^2 \) and \( \hat{J}^z \) with eigenvalues \( J(J + 1) \) and \( M \) respectively. Consider first the subspace with \( j_A = n_A I \) and \( j_B = (N - n_A) I \) where \( n_A \) and \( N - n_A \) are the number of nuclei of spin-\( I \) in group A and B respectively. \( j_A \) and \( j_B \) are conserved quantum numbers in the dynamics. Dicke states in this subspace can be uniquely specified as \( |J, M \rangle \).

The first non-trivial element for preparing Dicke states is the deterministic population transfer from \( |J, M + 1 \rangle \) to \( |J, M \rangle \). Only the first scenario for the phase control is needed where \( \hat{H}_c \) conserves the quantum number \( J \). We assume the population on electron excited state \( |e_0 \rangle \) can be efficiently damped to ground state with rate \( \Gamma \), which can be realized, e.g., via the tunneling process between the donor and the SET island at low temperature\(^{[23]} \). If the ac control frequency \( \omega \) is in resonance with the transition \( |g \rangle |J, M + 1 \rangle \leftrightarrow |e_0 \rangle |J, M \rangle \), the population on \( |g \rangle |J, M + 1 \rangle \) is pumped one-way to \( |g \rangle |J, M \rangle \) via the Raman-type process [Fig. 1(c)]. For efficient initialization of the electron spin on the ground state, the electron Zeeman energy shall be large as compared to the temperature. With a typical temperature of \( \sim 100 \text{ mK} \) used in the experiment\(^{[23]} \), a magnetic field \( \geq 0.3 \text{ T} \) is required, which corresponds to ac modulation of the A-gate voltage in the frequency range \( \geq 10 \text{ GHz} \).

There exists two unwanted couplings by the electron-nuclear flip-flop: (1) the coupling to other electron one-spin excitation states which are detuned at least by \( \Delta \) [Fig. 1(b)]; (2) the out-coupling of the final state \( |g \rangle |J, M \rangle \) to \( |e_0 \rangle |J, M - 1 \rangle \) which is also detuned by \( 2N a_0 \). For the latter, we note that for any pair of states \( |g \rangle |J, M + 1 \rangle \) and \( |e_0 \rangle |J', M \rangle \) coupled by \( \hat{H}_c \), the resonant frequency is \( \omega - \omega_N + a_0 + 2M a_0 \), namely, the pumping is \( M \)-selective with proper choice of the ac control frequency. When the detuning \( \delta \equiv \min \{ \Delta, \frac{2}{N} a_0 \} \gg \Gamma \), these unwanted couplings cause a negligible leakage \( \propto (\frac{\delta}{\Gamma})^2 \).

With the nuclear spins initialized on the unentangled polarized state \( |J = N I, M = N I \rangle \), the above simple control can deterministically prepare all symmetric Dicke states. Examples for preparing symmetric Dicke states of \( N = 20 \) \(^{31}\text{P} \) nuclear spins are simulated using the master equation

\[
\dot{\rho} = i[\rho, \hat{H}] - \frac{\Gamma}{2} \left( |e_0 \rangle \langle e_0 | \rho \rho \langle e_0 | - 2|g \rangle \langle e_0 | \rho \langle e_0 | g \rangle \right).
\]

The results are shown in Fig. 2. Clearly, the state preparation has the advantage that it is insensitive to the shape and area of the control pulse \( \Omega(t) \). We take \( a_0 / 2\pi = 60 \text{ MHz} \), \( \Gamma / 2\pi = 60 \text{ kHz} \), and assume the donor electron exchange of 0.5 GHz which leads to \( \Delta / 2\pi \sim 25 \text{ MHz} \). The preparation takes a time \( t_p \sim O(10) \mu s \). In comparison, a single two-qubit nuclear spin gate mediated by such a small donor exchange would take \( \sim 1 \text{ ms} \)^{[K]}. The probability to obtain each Dicke state is nearly unit (\( \geq 99.5\%) \), and the imperfection is caused by...
the off-resonance couplings which maybe further reduced by using smaller $\Omega$ with the cost of longer preparation time. We further note that the reverse population transfer from $|J, M\rangle$ to $|J, M + 1\rangle$ can be realized if global $\pi$ flips of nuclear spins are applied before and after the above pumping process with the central frequency set at $\omega = \omega_N + a_0 - (2(M + 1))/N\, a_0$.

The second non-trivial element is to realize the asymmetric Dicke state $|J = J_0, M = J_0\rangle$ with general values of $J_0$. This requires the quantum number $J_0$ to be changed which needs the control with the second scenario for phases. As discovered by the authors in our earlier work,[31,32] the inhomogeneous collective operator $\hat{J}_A - \hat{J}_B$ couples $|J, M + 1\rangle$ to the states $|J + \Delta J, M\rangle$ with the selection rule $\Delta J = 0, \pm 1$. Hence $\hat{H}_C$ can resonantly drive the one-way pumping from $|J, M + 1\rangle$ to $|J + \Delta J, M\rangle$ (see Fig. 1(d)). By repeating the above pumping for $N_1 > J_0$ steps, the population can be transferred from the initial state $|J = N_1, M = N_1\rangle$ to the states $|J \geq J_0, M = J_0\rangle$ (see Fig. 3(a)). If the reverse pumping from $|J, M = J_0\rangle$ to $|J, M = J_0 + 1\rangle$ (see proceeding paragraph) is also turned on, then all pathways from the initial state will end up at the desired target state $|J = J_0, M = J_0\rangle$ where the population gets trapped (see Fig. 3(a)). This realizes the deterministic preparation of $|J = J_0, M = J_0\rangle$ with arbitrarily specified $J_0$. Fig. 3(b) shows the numerical simulation for preparing the Dicke state $|J = 7, M = 7, J_A = 4, J_B = 6\rangle$ of 20 $^{31}$P nuclear spins, obtained with a probability of 99.5%.

Combining these two elements, an arbitrary Dicke state can be deterministically prepared in the subspace defined by $J_A = n_A I$ and $J_B = (N - n_A) I$. Essentially, this is the realization of an arbitrary total spin eigenstate for the two collective spins $\hat{J}_A$ and $\hat{J}_B$. By concatenating this procedure, we can deterministically prepare any state in a complete Dicke states basis for $N$ spins, denoted as $|J, M, \{j_{l,k}\}\rangle$ where $\{j_{l,k}\}$ are the collective spin of subsets [Fig. 4(a)]. The concatenated procedures are illustrated in Fig. 4(b).

![FIG. 4: Preparation of Dicke states $|J, M, \{j_{l,k}\}\rangle$ which spans a complete basis for the nuclear spin qubits. (a) The quantum numbers $j_{l,k}$ are the collective spin of subsets of qubits. (b) Concatenated preparation process. In step $l$, Dicke states are parallelly prepared in each 2$^l$-qubit subset with the collective spin being the specified value $j_{l,k}$. The quantum numbers $j_{l<0,k}$ are all conserved in step $l_0$. Red solid (black dashed) lines indicate the donor electron exchange ‘on’ (‘off’) when nuclear spins are pumped.]

III. EFFECTS OF POSSIBLE IMPERFECTIONS

Here we analyze effects of various imperfections that may exist in realistic systems and give recipes on how to deal with these sources of errors.

The first cause of error is the decoherence of the spin qubits. Dicke states are immune to decoherence in a collective form, but independent nuclear spin decoherence can cause a leakage out of the desired subspace. For a $N$-qubit state, the leakage is $\sim t_P N \gamma$ where $\gamma$ is the nuclear spin decoherence rate and $t_P$ is the preparation timescale proportional to $\Gamma^{-1}$. The longest nuclear spin coherence time reported for $^{31}$P donor in silicon is 1.75 second.[8] In Table 1, we show the performance of preparing several exemplary Dicke states in presence of spin decoherence. Nuclear spin pure dephasing described by the Lindblad term $-\gamma/2 \sum_n \left( \hat{I}_n^z \hat{I}_n^z \rho + \rho \hat{I}_n^z \hat{I}_n^z - 2 \hat{I}_n^z \hat{I}_n^z \right)$ is added to the master equation. The much slower nuclear spin relaxation process is neglected.[33] Because of the short preparation timescale, there is no visible effect from the decoherence if we take $\gamma/2\pi = 0.1$ Hz from the state-of-art measurements.[13] Even with the much exaggerated decoherence rate $\gamma/2\pi = 10$ Hz, the target states can still be obtained with high fidelity.

In realistic systems, there also exist various defects in the surrounding of the donors such as the interface $P_{00}$ centers.[10][34] The interplay between $P_{00}$ centers and phosphorus electron spins is important in the presence of photo-excited electrons and holes. Such interplay can facilitate the electrical detection of paramagnetic resonance of the donor electron.[10][34][35]. In the absence of illumination as in our scenario, at temperature of $k_B T \ll g \mu_B$, the $P_{00}$ center as a mid-gap paramagnetic center can lead to statistical fluctuations of the electron-nuclear flip-flop resonances from donor to donor. Furthermore, there could be noises from the local electrostatic environment, e.g. possible charge traps in the gate oxides[13] which may affect the hyperfine coupling and the donor electron exchange as well. If the charge hopping is faster than the state preparation timescale, the effect of this dynamics noise shall be similar to the effect of spin decoherence analyzed above. It is also possible to interlace the pumping control with sequence of $\pi$ pulses applied to the spin qubits for dynamical decoupling from such noises.[31] If charge hopping is much slower as compared to the state preparation timescale,
the noises are static which also result in the statistical fluctuation of the hyperfine coupling strength and exchange coupling strength.

To deal with such statistical fluctuations of system parameters, the system provides multi-fold individual tunability for each donor. The electron-nuclear flip-flop resonance depends on the static part of the hyperfine interaction (see Eq. (2a)) which can be independently controlled by each A-gate. Thus, in presence of inhomogeneous broadening of the electron-nuclear flip-flop resonances, the dc part of the A-gate voltage can tune the static part of the hyperfine coupling for compensating the inhomogeneity after calibration. As the excited state for the electron-nuclear flip-flop has an intrinsic broadening of $\Gamma$, inhomogeneity in the flip-flop resonances is unimportant if it is less than this intrinsic level broadening.

The exchange couplings between donor electrons are in general inhomogeneous due to the exchange oscillation problem and the possible effects of the aforementioned charge noises. This inhomogeneity manifests as nonuniform amplitudes $\alpha_n$ in the electron excited state $|\psi_0\rangle \equiv \sum_n \alpha_n \hat{\sigma}^+_n |g\rangle$. We note that the pumping operators that cooperatively flips may be flipped instead by $\hat{\sigma}^+_n$ due to the inhomogeneity in $\alpha$, strength $\delta V$, couplings and hence the values of $\alpha_n$. Then

$$\sum_n \frac{\partial \Omega}{\partial a_n} \delta V_n \alpha_n e^{i \phi_n} \hat{I}^-_n \quad \text{see Eq. (29).}$$

While J-gate voltages can tune the exchange couplings and hence the values of $\alpha_n$, the ac modulation strength $\delta V_n$ and phase $\phi_n$ can further compensate the remaining inhomogeneity in $\alpha_n$.

If only entanglement generation is of interest, our scheme can naturally cope with unknown systematic errors in the cooperative pumping. For controls aimed at the symmetric Dicke state $|J^-\rangle = |N/2, M = N/2\rangle$, the nuclear spins may be flipped instead by $\hat{A}^- \equiv \sum_n (1 + \eta_n) e^{i \theta_n} \hat{I}^-_n$ where $\theta_n$ and $\eta_n$ are respectively the unknown phase and amplitude errors. In presence of these errors, the steady state is still a definite pure state $|\psi_3\rangle = |\psi_3\rangle$ with similar multipartite entanglement. This feature makes the requirement less stringent for implementing this scheme for entanglement generation.

Moreover, one advantage of our scheme is unwanted donor sites can be easily disconnected in the state preparation. There are two ways to do this. One is to turn off the coupling of such donor site with others by the J-gate control. Even if the exchange interaction couples all neighboring donor electron spins into collective states, the set of nuclear spins being pumped can be further selected by the ac voltage controls. If the voltage applied to one of these donors has no ac component, its nuclear spin will not be pumped and is effectively decoupled from the rest in the preparation process. This advantage allows one to select out the set of donors where the parameters are relatively uniform, and it also facilitates the realization of the concatenated preparation process described in Fig. 4.

Finally, we give an estimate on the scale of the Dicke states that can be prepared using this approach. First, the detuning of the unwanted transitions is $\sim \frac{1}{4} \pi a_0$ which scales inversely with the qubit number $N$. To suppress the unwanted transitions, we require $\frac{1}{4} \pi a_0 \gg \Gamma$. Second, the leakage due to qubit decoherence is $\sim t_p N \gamma$, while the preparation time $t_p$ is determined by the smaller one of the modulation amplitude of the hyperfine interaction $\Omega$ and the spin initialization rate $\Gamma$. In our simulation, we take $\Gamma \sim 50 - 60$ kHz and $\Omega \sim 30 - 40$ kHz, both values in the range reported in experiments. Then $N$ shall not exceed 100 by the first requirement mentioned above. The upper bound for $N$ set by the second requirement depends on the qubit decoherence rate $\gamma$. For given $\gamma$, the fidelity for state preparation in a block of $N$ qubits may be scaled from the numbers listed in Table I. For example, with the nuclear spin decoherence rate of 10 Hz ($T_2 \sim 15$ ms), preparation of asymmetric Dicke state $|\psi_3\rangle$ for 20 qubits can have a fidelity over 0.96. Symmetric Dicke states may be prepared in a much larger scale, since they take much less steps by the cooperative pumping and $t_p$ is then considerably shorter.

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