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Coherence and Control of Quantum Registers Based on Electronic Spin in a Nuclear Spin Bath

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We consider a protocol for the control of few-qubit registers comprising one electronic spin embedded in a nuclear spin bath. We show how to isolate a few proximal nuclear spins from the rest of the bath and use them as building blocks for a potentially scalable quantum information processor. We describe how coherent control techniques based on magnetic resonance methods can be adapted to these solid-state spin systems, to provide not only efficient, high fidelity manipulation but also decoupling from the spin bath. As an example, we analyze feasible performances and practical limitations in the realistic setting of nitrogen-vacancy centers in diamond.

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The coherence properties of isolated electronic spins in the solid state are often determined by their interactions with large ensembles of lattice nuclear spins [1,2]. The nuclear spin dynamics is typically slow, resulting in very long bath correlation times. Indeed, nuclear spins are one of the most isolated systems in nature. This allows us to decouple electronic spins from nuclear spins via spin-echo techniques [3,4]. More remarkably, controlled manipulation of the coupled electron-nuclear system allows one to exploit the nuclear spin bath as a long-lived quantum memory [5–7]. Recently this approach has been used to prove a single nuclear qubit memory with coherence times exceeding tens of milliseconds in room temperature diamond [8]. Entangled states composed of one electronic and two nuclear spins have also been probed [9]. The essence of these experiments is to gain complete control over several nuclei by isolating them from the rest of the bath. Universal control of systems comprising a single electronic spin coupled to many nuclear spins has not been proved yet and could yield robust quantum nodes for larger scale quantum information architectures.

In this Letter we describe a technique to achieve universal control of a portion of the nuclear spin bath and use it, together with an electronic spin, to build multiqubit registers. Our approach is based on magnetic resonance control techniques, but there is an essential difference with previously studied small quantum processors, such as NMR molecules. Here the boundary between qubits in the system and bath spins is ultimately dictated by our ability to effectively control the qubits. The interactions between the electronic spin and the nuclear qubit and bath spins are of the same nature, so that control schemes must preserve the desired interactions among qubits while attempting to remove the couplings to the bath. The challenge to overcome is then to resolve individual energy levels for qubit addressability and control, while avoiding fast dephasing due to the uncontrolled portion of the bath.

Before proceeding we note that proposals for integrating small quantum registers into a larger system capable of fault tolerant quantum computation or communication have been explored theoretically [10,11] and experimentally [12,13]. These schemes generally require a *communication* qubit and a few *ancillary* qubits per register. The first one couples efficiently to external degrees of freedom (for initialization, measurement and entanglement distribution), leading to easy control but also fast dephasing. The *ancillary* qubits instead are more isolated and act as memory or ancillas in error correction protocols. While our analysis is quite general as it applies to various systems, e.g., quantum dots in carbon nanotubes [14] or spin impurities in silicon [7], we will focus on the nitrogen-vacancy (NV) centers [4,8,9] in high-purity diamond with low paramagnetic impurity content. These are promising systems to implement hybrid quantum networks due to their long coherence times and good optical transitions that can be used for remote coupling among registers, without overly affecting the coherence of the register's ancillary qubits [15]. Specifically, nuclear spins can remain coherent for a large number of electronic excitation cycles [8].

We focus on an electronic-spin triplet, as for the NV centers (see Fig. 1). Quantum information is encoded in the $m_s = \{0, 1\}$ Zeeman sublevels, separated by a large zero field splitting (making m_s a good quantum number). Other Zeeman levels are shifted off resonance by an external magnetic field B_z along the N-to-V axis. The Hamiltonian in the electronic rotating frame is

$$\begin{aligned} \mathcal{H} &= \omega_L \sum I_z^j + S_z \sum A_j \cdot \vec{I}^j + \mathcal{H}_{\text{dip}} \\ &= \frac{\mathbb{1} - S_z}{2} \omega_L \sum I_z^j + \frac{\mathbb{1} + S_z}{2} \sum \tilde{\omega}_1^j \cdot \vec{I}^j + \mathcal{H}_{\text{dip}}, \end{aligned} \quad (1)$$

where S and I^j are the electronic and nuclear spin operators, ω_L the nuclear Larmor frequency, A_j the hyperfine

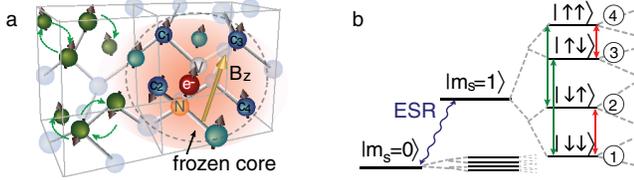


FIG. 1 (color online). (a) System model, showing the electronic spin in red and surrounding nuclear spins. The closest nuclear spins are used as qubits. Of the bath spins, only the ones outside the frozen core evolve due to dipolar interaction, causing decoherence. (b) Frequency selective pulses, in a 3-qubit register.

couplings and \mathcal{H}_{dip} the nuclear dipolar interaction, whose strength can be enhanced by the hyperfine interaction [8]. When the electronic spin is in the $m_s = 1$ state, nearby nuclei in the so-called frozen core [16] are static (since distinct hyperfine couplings make nuclear flip-flops energetically unfavorable) and give rise to a quasistatic field acting on the electronic spin. Other bath nuclei cause decoherence via spectral diffusion [17], but their couplings, setting noise strength and correlation time, are orders of magnitude lower than the interactions used for control. While in the $m_s = 0$ manifold all the nuclear spins precess at the same frequency, the effective frequencies in the $m_s = 1$ manifold, ω_1^j , are given by the hyperfine interaction and the enhanced g tensor [18], yielding a wide range of values. Some of the nuclear spins in the frozen core can thus be used as qubits.

Control is obtained via microwave (μw) and radio frequency (rf) fields. The most intuitive scheme, performing single-qubit gates with these fields and two-qubit gates by direct spin-spin couplings, is quite slow, since rf transitions are weak. Another strategy, requiring only control of the electronic spin, has been proposed [19,20]: switching the electronic qubit between its eigenstates induces nuclear spins rotations about two nonparallel axes that generate any single-qubit gate. This strategy is not the most appropriate here, since rotations in the $m_s = 0$ manifold are slow [21]. We thus propose another scheme to achieve universal control, using only two types of gates: (i) One-qubit gates on the electronic spin and (ii) Controlled gates on each of the nuclear spins. The first gate is simply obtained by a strong μw pulse. The controlled gates are implemented with rf pulses on resonance with the effective frequency of individual nuclear spins in the $m_s = 1$ manifold, which are resolved due to the hyperfine coupling and distinct from the bath frequency [22]. Achievable rf power provides fast enough rotations since the hyperfine interaction enhances the nuclear Rabi frequency when $m_s = 1$ [23]. Any other gate needed for universal control can be obtained combining these two gates. For example, we achieve a single nuclear qubit rotation by repeating the controlled gate after applying a π -pulse to the electronic spin. Controlled gates between nuclei can be implemented by exploiting the stronger coupling to the electron as shown in Fig. 2(a).

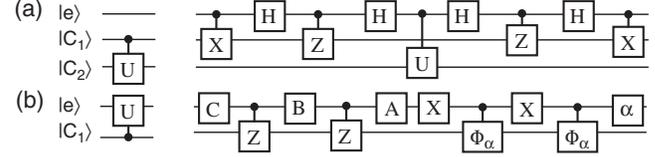


FIG. 2. Circuits for controlled gates U among two nuclear spins (a) and a nuclear and the electronic spin (b). The gates A , B , C are defined such that $U = e^{i\alpha}AZBZC$ and $ABC = \mathbb{1}$, where Z is a π rotation around z [35]. Φ_α is a phase gate: $|0\rangle \times \langle 0| + |1\rangle \langle 1| e^{i\alpha/2}$ and the gate α indicates $e^{i\alpha/2} \mathbb{1}$.

Avoiding direct nuclear interactions is faster as long as the hyperfine coupling is several times larger than the nuclear coupling.

Although selectively addressing ESR transitions is a direct way to perform a controlled rotation with the electronic spin as target, this is inefficient as the number of nuclear spin increases. The circuit in Fig. 2(b) performs the desired operation on a faster time scale.

When working in the $m_s = 1$ manifold, each nuclear qubit is quantized along a different direction and we cannot define a common rotating frame. The evolution must be described in the *laboratory* frame while the control Hamiltonian is fixed in a given direction for all the nuclei (e.g., the x axis). This yields a reduced rf Rabi frequency $\tilde{\Omega} = \Omega_{\text{rf}} \sqrt{\cos\varphi_1^2 \cos^2\theta_1^2 + \sin\varphi_1^2}$ (where $\{\theta_1, \varphi_1\}$ define local quantization axes in the $m_s = 1$ manifold and Ω_{rf} is the hyperfine-enhanced rf frequency). The propagator for a pulse time t_p and phase ψ is

$$U_L(\Omega_{\text{rf}}, t_p, \psi) = e^{-i[\omega t_p - (\lambda - \psi)]\sigma_z/2} e^{-i\tilde{\Omega} t_p \sigma_x/2} e^{-i(\lambda - \psi)\sigma_z/2},$$

where $\{\sigma_{\tilde{x}}, \sigma_{\tilde{y}}, \sigma_{\tilde{z}}\}$ are the Pauli matrices in the local frame and $\tan(\lambda) = \tan\varphi_1 / \cos\theta_1$. An arbitrary gate $U = R_{\tilde{z}}(\gamma)R_{\tilde{x}}(\beta)R_{\tilde{z}}(\alpha)$ is obtained by combining U_L with an echo scheme (Fig. 3), which not only refocuses the extra free evolution due to the lab frame transformation, but also sets the gate duration to any desired clock time. Fixing a clock time common to all registers is advantageous to synchronize their operation.

In order to refocus the fast electronic-spin dephasing due to the frozen core nuclear spins, we need to embed the control strategy described above in a dynamical decoupling scheme [24] without losing universal control. The electron-bath Hamiltonian is given by Eq. (1), where the index j now runs over the bath spins. Neglecting for now

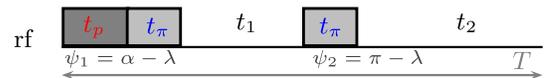


FIG. 3 (color online). rf pulse scheme for 1 nuclear spin gate in the $m_s = 1$ manifold. With $t_p = \beta/\tilde{\Omega}$ and fixing $\psi_1 = \alpha - \lambda$ and $\psi_2 = \pi - \lambda$, the time delays are $t_1 = \frac{T}{2} - t_p - t_\pi - \frac{\alpha + \gamma}{\omega}$ and $t_2 = \frac{T}{2} - t_\pi + \frac{\alpha + \gamma}{\omega}$. This yields a minimum clock time $T \geq 4\pi \text{Max}_{\tilde{\Omega}_j, \omega_j^i} \{\tilde{\Omega}_j^{-1} + (\omega_j^i)^{-1}\}$.

the couplings among nuclei, we can solve for the electronic-spin evolution under an echo sequence. By defining U_0 and U_1 the propagators in the 0 and 1 manifolds and assuming the nuclear spins initially in the identity state (high temperature regime), we calculate the dynamics of the electronic spin, $\rho_e(t) = [\mathbb{1} + (|0\rangle\langle 1|f_{ee}(t) + \text{H.c.})]\rho_e(0)$, where $f_{ee}(t) = \text{Tr}[U_1 U_0 U_1^\dagger U_0^\dagger] = \prod [1 - 2\sin^2(\theta_1^j)\sin^2(\omega_1^j t/2)\sin^2(\omega_L t/2)]$ is the function describing the echo envelope experiments [18,25]. Since in the $m_s = 0$ subspace all the spins have the same frequency, $f_{ee}(2n\pi/\omega_L) = 0$ and the electron comes back to the initial state. Nuclear spin-spin couplings lead to imperfect echo revival and ultimately to decoherence via spectral diffusion [17]. The energy-conserving flip-flops of remote nuclear spins modulate the hyperfine couplings, causing the effective field at the electron to vary in time. The field oscillations can be modeled by a classical stochastic process. The evolution of the electronic spin is thus due to two processes that can be treated separately as they commute: the echo envelope calculated above and the decay due to a stochastic field, approximated by a cumulant expansion [26]. For Lorentzian noise with autocorrelation $G(\tau) = G_0 e^{-(t/\tau_c)}$ we obtain a spin-echo decay $\propto e^{-(2\Omega^2/3\tau_c)t^3}$ for $t \ll \tau_c$. By using dynamical decoupling techniques [27] and selecting a cycle time multiple of the Larmor precession period it is possible to prolong the electronic coherence. Figure 4 shows how to combine the electron modulation with the sequence implementing spin gates. The effectiveness of these techniques relies on the ability to modulate the evolution on a time scale shorter than the noise correlation time. The noise of the electron-nuclear spin system is particularly adapt to these decoupling schemes. Consider, for example, the NV center. The measured electron dephasing T_2^* time is about $1 \mu\text{s}$ in natural diamonds [18], as expected from MHz-strong hyperfine couplings. The intrinsic decoherence time T_2 can be orders of magnitude longer ($T_2 \gtrsim 600 \mu\text{s}$), reflecting the existence of the frozen core, where spin flip flops are quenched. The frozen core radius is about 2.2 nm [16] and only spins with hyperfine coupling $\lesssim 2.5 \text{ kHz}$ contribute to spectral diffusion. Both the inverse correlation time (given by the nuclear dipolar coupling) and the noise rms (given by the coupling to the electron) are of order of few kHz. Dynamical decoupling schemes must thus act on time scales of hundreds of μs . This in turns sets achievable constraints on the gate speed.

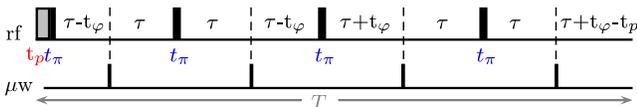


FIG. 4 (color online). rf and μw pulse sequence to implement a 1 nuclear spin gate while reducing the effects of a slowly varying electronic dephasing noise. The black bars indicate π -pulses, while the first rectangle indicate a general pulse around the x axis. $\tau = T/8 - t_\pi/2$ and $t_\phi = (\alpha + \gamma)/4\omega$.

The time of a conditional single nuclear spin rotation must be set so that Ω_{rf} is much less than the frequency splitting between two *neighboring* spins (in terms of frequencies). Suppose we want to control an n -spin register without exciting bath spins at the Larmor frequency. The minimum frequency splitting between two nuclei in the $m_s = 1$ manifold will be at best $\delta\omega = \frac{\omega_M}{n}$ for nuclear frequencies equally spaced and ω_M the maximum hyperfine-induced effective nuclear frequency. The nuclear frequency spread due to the hyperfine interaction is then $\Delta E_N = \frac{n+1}{2}\omega_M$. We want the Rabi frequencies to satisfy the constraints: $\Delta E_e \gg \Omega_e \gg \frac{n+1}{2}\omega_M > \frac{\omega_M}{n} \gg \Omega_{\text{rf}}$, where $\Delta E_e = 2g\mu_B B_z$ is the gap to other Zeeman levels ($m_s = -1$ for the NVc) and Ω_e the μw power. For a typical choice of 700 G magnetic field, we have $\Delta E_e = 2 \text{ GHz}$ and $\omega_L = 0.8 \text{ MHz}$. Assuming, $\omega_M \approx 20 \text{ MHz}$ and $n = 4$ spins, we obtain the following parameter window (in MHz) $2000 \gg \Omega_e \gg 25, \Omega_{\text{rf}} \ll 5$. The gate clock time can be as short as a few μs , allowing hundreds of gates in the coherence time.

Since the scheme presented is based on selective pulses, the dominant (coherent) error is due to off-resonance effects. If the Rabi frequency is much smaller than the offset from the transmitter frequency $\delta\omega$, the off-resonance spin will just experience a shift (Bloch-Siegert shift) of its resonance frequency, $\Delta\omega_{\text{BS}} = \delta\omega - \frac{1}{t} \int \sqrt{\Omega_{\text{rf}}(t)^2 + \delta\omega^2} dt' \approx -\frac{\Omega_{\text{rf}}^2}{2\delta\omega} = -\frac{n\Omega_{\text{rf}}^2}{2\omega_M}$. This results in an additional phase acquired during the gate time that needs to be refocused. Note that this error grows with the register size and constrains Ω_{rf} . When reducing the Rabi frequency to achieve frequency selectivity, we have to pay closer attention to the rotating-wave approximation and consider its first order correction, which produces a shift of the on-resonance spin $\Delta\omega_{\text{RWA}} = \Omega_{\text{rf}}^2/4\omega_M$. Other sources of errors are evolution of bystander nuclear spins and couplings among spins. More complex active decoupling schemes [28,29] can correct for these errors, allowing to use the $m_s = 1$ manifold as a memory.

Advanced techniques like shaped pulses, with amplitude and phase ramping, composite pulses [30], optimal control theory or numerical techniques [31,32] can provide better fidelity. The analytical model of control serves then as an initial guess for the numerical searches. Pulses found in this way correct for the couplings among nuclei and are robust over a wide range of parameters (such as experimental errors or the noise associated with static fields). Table I shows the results of simulations in a fictitious NV system with 1–4 nuclear spins and effective frequencies in the $m_s = 1$ manifold ranging from 15 to 2 MHz. We searched numerically via a conjugate gradient algorithm for a control sequence performing a desired unitary evolution, varying amplitude and phase of the μw and rf fields. We then simulated the control sequence in the presence of noise, with contributions from a large, static field and a smaller fluctuating one. The projected fidelities in the

TABLE I. Simulated fidelities ($F = |\text{Tr}[U_w^\dagger U]/2^N|^2$) at the optimal gate time. The gate is a $\pi/2$ rotation about the local x axis for qubit 1 in a register of 1–4 nuclear qubits. Noise parameters are $T_2^* = 1.5 \mu\text{s}$ and $T_2 = 250 \mu\text{s}$; maximum $\mu\omega$ and rf Rabi frequencies are $2\pi \times 10$ MHz and $2\pi \times 20$ kHz. As expected, the optimal gate time increases with register size, reflecting both more complex control required in a larger Hilbert space and weaker hyperfine couplings to distant spins. We obtained similar results for a CNOT gate between spin 1 and 2.

	1 spin	2 spins	3 spins	4 spins
F (ideal)	0.9999	0.9999	0.9992	0.9995
F (noise)	0.9994	0.9995	0.9975	0.9925
time	$5.0 \mu\text{s}$	$5.5 \mu\text{s}$	$6.0 \mu\text{s}$	$8.5 \mu\text{s}$

absence of experimental errors are very high, a sign that fast modulation of the electronic spin effectively decouples it from the bath. Pulse robustness to noise is slightly degraded as the spin number increases: as the noise induces a spread of the electron frequency, it becomes difficult to find a sequence of control parameters that drives the desired evolution in the larger Hilbert space for every electronic frequency. The fidelity degradation is still modest and can be in part corrected by increasing the control field intensity. Combining these pulses with dynamical decoupling would provide an efficient way to coherently control the registers.

The register size is eventually limited by the number of nuclear spins with hyperfine coupling strong enough to be separated from the bath. From experiments and *ab initio* calculation [33] we expect hyperfine couplings of ~ 130 MHz in the first shell, and then about 50 possible nuclear sites with hyperfine values from 15 to 1 MHz. Even if the concentration of ^{13}C is increased (and the nitrogen nuclear spin used) the size of the register will be limited to about 10 spins. Still, such registers would be very useful for memory and error correction purposes.

In conclusion, we have presented a general approach to the control of small quantum systems comprising an electronic spin and few nuclear spins in the surrounding spin bath. We have shown that several of the bath spins can be isolated and effectively controlled, yielding a few-qubit register. These registers can be used in proposals for distributed quantum computation and communication, where coupling among registers could be provided either via photon entanglement [15] or by a movable reading tip [34]. Our control methods enable algorithms needed for error correction and entanglement purification, while the nuclear spins provide a longtime memory in the $m_s = 1$ manifold, via active refocusing, and the electron dephasing is kept under control by dynamical decoupling methods. We thus develop a modular control scheme, scalable to many registers and applicable to various physical implementations.

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