Suppressing Spin Qubit Dephasing by Nuclear State Preparation

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Coherent spin states in semiconductor quantum dots offer promise as electrically controllable quantum bits (qubits) with scalable fabrication. For few-electron quantum dots made from gallium arsenide (GaAs), fluctuating nuclear spins in the host lattice are the dominant source of spin decoherence. We report a method of preparing the nuclear spin environment that suppresses the relevant component of nuclear spin fluctuations below its equilibrium value by a factor of ~70, extending the inhomogeneous dephasing time for the two-electron spin state beyond 1 \( \mu s \). The nuclear state can be readily prepared by electrical gate manipulation and persists for > 10 s.

Quantum information processing requires the realization of interconnected, controllable quantum two-level systems (qubits) that are sufficiently isolated from their environment that quantum coherence can be maintained for much longer than the characteristic operation time. Electrons spins in quantum dots are an appealing candidate system for this application, as the spin of the electron is typically only weakly coupled to the environment compared to the charge degree of freedom \([4]\). Logical qubits formed from pairs of spins provide additional immunity from collective dephasing, forming a dynamical decoherence-free subspace \([2, 3]\).

Implementing any spin-qubit architecture requires the manipulation \([4, 5, 6]\) and detection \([7, 8]\) of few-electron spin states, as yet only demonstrated in III-V semiconductor heterostructure devices such as gallium arsenide (GaAs), which, in all cases, comprise atoms with nonzero nuclear spin. The nuclear spins of the host lattice couple to electrons via the hyperfine interaction and causes rapid electron spin dephasing. In the GaAs devices presented here, for instance, an ensemble of initialized spin pairs will retain their phase relationship for \( T_s \approx 10 \) ns, consistent with theoretical estimates \([9, 10, 11]\) and previous measurements \([4]\). The time \( T_s \) represents an inhomogeneous dephasing time, which can be extended using spin-echo methods \([4]\). Extending \( T_s \) by nuclear state preparation reduces the burden of using echo sequences or large field gradients to overcome the fluctuating hyperfine fields when controlling spin qubits.

Proposals to reduce dephasing by nuclear state preparation include complete nuclear polarization \([12]\), state-narrowing of the nuclear distribution \([12, 13, 14, 15]\), and schemes for decoupling the bath dynamics from the coherent evolution of the electron spin using control pulses \([16, 17, 18]\). These approaches remain largely unexplored experimentally, though recent optical experiments \([19]\) have demonstrated a suppression of nuclear fluctuations in ensembles of self-assembled quantum dots.

We demonstrate a nuclear state preparation scheme in a double quantum-dot system using an electron-nuclear flip-flop pumping cycle controlled by voltages applied to electrostatic gates. Cyclic evolution of the two-electron state through the resonance between the singlet \((S)\) and \( m_s = 1 \) triplet \((T_+)\) \([20]\) leads to a 70-fold suppression of fluctuations below thermal equilibrium of the hyperfine field gradient between the dots along the total field direction. It is this component of the hyperfine field gradient that is responsible for dephasing of the two-electron spin qubit formed by \( S \) and \( m_s = 0 \) triplet \((T_0)\) states \([3]\). Consequently, though the flip-flop cycle generates only a modest nuclear polarization (< 1%), the resulting nuclear state extends \( T_s \) of the \( S - T_0 \) qubit from 15 ns to beyond 1 \( \mu s \). Once prepared, this non-equilibrium nuclear state persists for ~15 s, eventually recovering equilibrium fluctuations on the same time scale as the relaxation of the small induced nuclear polarization. Noting that this recovery time is ~9 – 10 orders of magnitude longer than typical gate operation times, we propose that occasional nuclear state preparation by these methods may provide a remedy to hyperfine-mediated spin dephasing in networks of interconnected spin qubits.

The double quantum dot is defined in a GaAs/AlGaAs heterostructure with a two-dimensional electron gas (2DEG) 100 nm below the wafer surface (density \( 2 \times 10^{15} \text{m}^{-2} \), mobility \( 20 \text{m}^2/\text{Vs} \)). Negative voltages applied to Ti/Au gates create a tunable double-well potential that is tunnel coupled to adjacent electron reservoirs \([21]\). A proximal radio-frequency quantum point contact (rf-QPC) senses the charge state of the double dot, measured in terms of the rectified sensor output voltage \( V_{rf} \) \([22]\). Measurements were made in a dilution refrigerator at base electron temperature of 120 mK.

A schematic energy level diagram (Fig. 1A), with \((n,m)\) indicating equilibrium charge occupancies of the left and right dots, shows the three \((1,1)\) triplet states \((T_+, T_0, T_-)\) split by a magnetic field \( B_0 \) applied perpendicular to the 2DEG. The detuning from the \((2,0)-(1,1)\) degeneracy, \( \varepsilon \), is controlled by high-bandwidth gate voltages pulses. The ground state of \((2,0)\) is a singlet, with the \((2,0)\) triplet out of the energy range of the experiment.

Each confined electron interacts with \( N \sim 10^6 \) nu-
local effective fields, which can be decomposed into an
average field and a difference field. It is useful to re-
solve $B_n = (B^1_n + B^2_n)/2$, the Overhauser part of
the total average field, $B_{\text{tot}} = B_0 + B_n$, into components
along $(B^1_n)$ and transverse $(B^2_n)$ to $B_{\text{tot}}$. The difference
field, due only to Overhauser contributions, is given by
$\Delta B_n = (B^1_n - B^2_n)/2$, with components along $(\Delta B^1_n)$
and transverse $(\Delta B^2_n)$ to $B_{\text{tot}}$. At large negative $\varepsilon$, where
the two electrons are well separated and exchange $J(\varepsilon)$ is
negligible, $\Delta B^1_n$ sets the precession rate between $S$ and $T_0$ states.
At the value of detuning where $J(\varepsilon)$ equals the
Zeeman energy $E_Z = g \mu_B B_0$, ($g$ is the electron $g$-factor
and $\mu_B$ is the Bohr magneton) precession between $S$ and $T_+$ states occurs at a rate set by $\Delta B^1_n$.

To measure the precession or dephasing of spin pairs
in the two dots, a gate-pulse cycle (“probe cycle”) first
prepares (P) a singlet state in (2,0), then separates (S)
the two electrons into (1,1) for a duration $\tau_S$, then mea-
sures (M) the probability of return to (2,0). States that
evolve into triplets during $\tau_S$ remain trapped in (1,1) by
the Pauli blockade, and are detected as such by the rf-
QPC charge sensor [4]. Figures 1C and D show the time-
averaged charge sensing signal, $V_{\text{rf}}$, as a function of con-
stant offsets to gate biases $V_L$ and $V_R$, with this pulse
sequence running continuously. Setting the amplitude of
the S-pulse to mix $S$ with $T_0$ at large detuning (green
dashed line, Fig. 1B) yields the “readout triangle” indi-
cated in Fig. 1C. Within the triangle, $V_{\text{rf}}$ is between (2,0)
and (1,1) sensing values, indicating that for some probe
cycles the system becomes Pauli blockaded in (1,1) after
evolving to a triplet state. Outside this triangle, alter-
native spin-independent relaxation pathways circumvent
the blockade [23]. For a smaller amplitude S-pulse (red
dashed line, Fig. 1B), $S$ mixes with $T_+$, also leading to
partial Pauli blockade and giving the narrow resonance
feature seen in Fig. 1D. The dependence of the $S - T_+$
resonance position on applied field $B_0$ serves as a cali-
bration, mapping gate voltage $V_L$ (at fixed $V_R$) into total
effective field, $B_{\text{tot}}$, including possible Overhauser fields
(Fig. 2A). The charge sensing signal, $V_{\text{rf}}$, is also cali-
brated using equilibrium (1,1) and (2,0) sensing values
in order to give the probability 1 $- P_S$ that an initialized singlet
will evolve into a triplet during the separation time $\tau_S$
(Fig. 2G). A fit to $P_S(\tau_S)$ (Fig. 2C) yields [11] [22] [24]
a dephasing time $T'_{2} = h/(g\mu_B^2)\langle\Delta B^1_{\text{rms}}\rangle$ $\sim$ 15 ns,
where $\langle...\rangle_{\text{rms}}$ denotes a root-mean-square-time-ensemble
average.

We now investigate effects of the electron-nuclear flip-
flop cycle (“pump cycle”) (Fig. 1E). Each iteration of the
pump cycle moves a singlet, prepared in (2,0), adiabat-
ically through the $S - T_+$ resonance, then returns non-
adabatically to (2,0), where the state is re-initialized to
a singlet by exchanging an electron with the adjacent
reservoir [20]. In principle, with each iteration of this
cycle, a change in the angular momentum of the electron
state occurs with a corresponding change to the nuclear
system. Iterating the pump cycle at 4 MHz creates a
modest nuclear polarization of order 1%, as seen previ-

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**FIG. 1:** (A) Schematic of the energy levels of the two-electron
system in a magnetic field. Detuning, $\varepsilon$, from the (2,0)-(1,1)
charge degeneracy is gate controlled. (B) Gate-pulse sequence
used to separately probe the longitudinal, $\Delta B^1_n$ (green dashed
line), and transverse, $\Delta B^2_n$ (red dashed line) components
of the Overhauser field difference, depending on the position of
the separation point $S$. (C and D) Time-averaged charge-
sensing signal, $V_{\text{rf}}$, from the rf-QPC, as a function of gate
bias $V_L$ and $V_R$, showing features corresponding to the sin-
glet mixing with the $T_0$ (bracketed green triangle in C) and
$T_+$ (bracketed green line segment in D). (E) Schematic view of
the $S - T_+$ anticrossing, illustrating the pumping cycle. With
each iteration of this cycle, with period $\tau_C = 250$ ns, a new
singlet state is taken adiabatically through the $S - T_+$ anti-
crossing in a time $\tau_A = 50$ ns, then returned nonadiabatically
to (2,0) in $\sim$ 1 ns, where the $S$ state is then reloaded.
The pump cycle was always iterated for more than 1 s, and no dependence on pumping time beyond 1 s was observed. What limits the efficiency of the pumping cycle, keeping the polarization in the few-mT regime, is not understood.

Immediately following the pump cycle, the gate-voltage pattern is switched to execute one of two types of probe cycles. The first type of probe cycle starts in (2,0) and makes a short excursion deep into (1,1) to measure $P_S$ (using data from Fig. 3A). The observed increase in $T_2^*$, slices through the readout triangle (as in Figs. 2E) are sampled as a function of time following pumping, calibrated using the same probe cycle without the preceding pump cycle, shown in Figs. 2H-I, do not show suppressed mixing of the separated singlet state.

Measurement of $P_S(\tau_S)$ as a function of $\tau_S$, slices through the readout triangle (as in Figs. 2E) are sampled as a function of time following pumping, calibrated using the out-of-triangle background, and averaged, giving traces such as those in Figs. 3B-E. Gaussian fits yield $T_2^* \sim 1 \mu s$ for 0-5 s after the pump cycle and $T_2^* \sim 0.5 \mu s$ for 5-10 s after the pump cycle. After 50 s, no remnant effect of the pump cycle can be seen, with $T_2^*$ returning to $\sim 15$ ns, as prior to the pump cycle.

The rms amplitude of longitudinal Overhauser field difference, $\langle \Delta B^*_n \rangle_{\text{rms}} = \hbar / g \mu_B T_2^* \bar{\tau}$, is evaluated using $T_2^*$ values within several time blocks following the pump cycle (using data from Fig. 3A). The observed increase in $T_2^*$ following the pump cycle is thus recast in terms of a suppression of fluctuations of $\Delta B^*_n$ (Fig. 4A).

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the $S - T_+$ mixing rate is used to infer the size of fluctuations of the transverse component of the Overhauser field, $\langle \Delta B_{n}^\perp \rangle_{\text{rms}}$. Figure 4B shows $P_S(\tau_S = 25 \mu s)$ near the $S - T_+$ resonance. Unlike $S - T_0$ mixing, which is strongly suppressed by the pump cycle, the $S - T_+$ resonance appears as strong as before the pump cycle. This suggests that the energy gap $E_\parallel^0$ (Fig. 4E) is not closed by the pump cycle. Note that fluctuations in $\Delta B_{n}^\perp$ produce fluctuations in $E_\parallel^0$, which give the $S - T_+$ anticrossing a width in detuning $\varepsilon$ (Fig. 4E). Converting to a width in magnetic field via Fig. 2(A), gives the fluctuation amplitude $\langle \Delta B_{n}^\perp \rangle_{\text{rms}}$ following the pump cycle. Figure 4C shows a representative slice taken from Fig. 4B at the position indicated by the white dashed line. Gaussian fits to each 1 s slice yield mean positions, $m$, and widths, $w$, in magnetic field that fluctuate in time, as seen in Fig. 4D. The increase in $w$ for short times ($t < 10$ s) reflects gate-voltage noise amplified by the saturating conversion from gate voltage to effective field at large $B_{\text{tot}}$ (see SOM). Beyond these first few seconds, $w$ is dominated by fluctuations of $\Delta B_{n}^\parallel$, but is also sensitive to fluctuations in $m$ that result from fluctuations of $B_{n}^\parallel$ (see Fig. 4E). (For $t > 10$ s, gate-voltage noise makes a relatively small (< 10 %) contribution to the fluctuations.) Estimating and removing the contribution due to $B_{n}^\parallel$ (see SOM) gives an estimate of $\langle \Delta B_{n}^\perp \rangle_{\text{rms}}$ as a function of time following the pump cycle. These results are summarized by comparing Fig. 4A and 4F: in contrast to the strong suppression of fluctuations in $\Delta B_{n}^\parallel$ following the pump cycle, no corresponding suppression of $\langle \Delta B_{n}^\perp \rangle_{\text{rms}}$ is observed.

Reducing the cycle rate by a factor $\sim 10$ reduces, but does not eliminate, the suppression of fluctuations of $\Delta B_{n}^\parallel$ remains evident (see SOM for discussion of dependence of polarization on pump cycle rate). Also, when the pump cycle is substituted by a cycle that rapidly brings the singlet into resonance with $T_0$, deep in (1,1), effectively performing multiple fast measurements of $\Delta B_{n}^\parallel$, no subsequent effect on $S - T_+$ mixing is observed. This demonstrates that it is transitions involving $S$ and $T_+$, rather than $S$ and $T_0$, that lead to the suppression of nuclear field gradient fluctuations.

The observation that an adiabatic electron-nuclear flip-flop cycle will suppress fluctuations of the nuclear field gradient has been investigated theoretically [25, 26]. These models explain some but not all of the observed phenomenology described here, and it is fair to say that a complete physical picture of the effect has not yet emerged. Other nuclear preparation schemes arising from various hyperfine mechanisms, not directly related to the specific pump cycle investigated here, have also been addressed theoretically in the recent literature [27, 28].

Control of spin qubits in the presence of time-varying equilibrium Overhauser gradients require complex pulse sequences [4] or control of sizable magnetic field gradients [2] [29]. Suppressing fluctuations of $\Delta B_{n}^\parallel$ by a factor of order 100, as demonstrated here using nuclear state preparation, leads to an improvement in control fidelity of order $10^4$, assuming typical control errors, which scale as $(\Delta B_{n}/\Delta B_{n}^\parallel)^2$ for low-frequency noise. We further anticipate generalizations of the present results using more than two confined spins that allow arbitrary gradients in nuclear fields to be created by active control of Overhauser coupling.
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