Measurement of Temporal Correlations of the Overhauser Field in a Double Quantum Dot

D. J. Reilly, J. M. Taylor, E. A. Laird, J. R. Petta, C. M. Marcus, M. P. Hanson and A. C. Gossard

1 Department of Physics, Harvard University, Cambridge, MA 02138, USA
2 Department of Physics, Massachusetts Institute of Technology, Cambridge, MA 02139, USA
3 Department of Physics, Princeton University, Princeton, NJ 08544, USA
4 Department of Materials, University of California, Santa Barbara, California 93106, USA

In quantum dots made from materials with nonzero nuclear spins, hyperfine coupling creates a fluctuating effective Zeeman field (Overhauser field) felt by electrons, which can be a dominant source of spin qubit decoherence. We characterize the spectral properties of the fluctuating Overhauser field in a GaAs double quantum dot by measuring correlation functions and power spectra of the rate of singlet-triplet mixing of two separated electrons. Away from zero field, spectral weight is concentrated below 10 Hz, with $\sim 1/f^2$ dependence on frequency, $f$. This is consistent with a model of nuclear spin diffusion, and indicates that decoherence can be largely suppressed by echo techniques.

Electron spins in quantum dots are an attractive candidate for quantum bits (qubits) [1, 2]. For gate-defined devices made using GaAs, the coupling of single electron spins to $\sim 10^6$ thermally excited nuclear spins creates a fluctuating effective Zeeman field (the Overhauser field), $B_{\text{nuc}}$, with rms amplitude $B_{\text{nuc}} \sim 1-3 \text{ mT}$ [3, 4, 5, 6, 7]. At experimentally accessible temperatures, $B_{\text{nuc}}$ fluctuates both as a function of position and time, with temporal correlations over a broad range of time scales, and is a dominant source of spin dephasing. Experiments have been performed both in GaAs [8, 9, 10, 11, 12, 13] and low-field spin relaxation [3, 4, 5, 6, 7]. Spin manipulation schemes [10, 11, 12, 13] can be employed to control spin dephasing, which can be observed and characterized using magnetometry [8, 9]. The conductance $G_{\text{rf}}$ of a proximal radio frequency quantum point contact (rf-QPC) is sensitive to the charge configuration of the double dot. $G_{\text{rf}}$ is sensitive to the charge degeneracy determined by electrostatic gates. A gate-pulse (Fig. 1(b)) cycle prepares new singlets each iteration by configuring the device deep in (2,0), at point (P), where transitions to the ground state singlet, (2,0)S, occur rapidly [9, 13]. Electrons are then separated to position S in (1,1) for a time $\tau_{\text{S}}$ where precession between the initial singlet and one of the triplet states is driven by components of the difference in Overhauser fields in the left and right dots, $\Delta B_{\text{nuc}} = B_{\text{nuc}}^{L} - B_{\text{nuc}}^{R}$ [9, 13].

In an applied field, the position of the separation point determines whether the (1,1) singlet $(S)$ is nearly degenerate with one of the (1,1) triplets, with which it can then rapidly mix. Mixing of $S$ with $T_0$ (the $m_s = 0$ triplet) occurs at large negative $\epsilon$ (green line in Fig. 1(b)) where exchange vanishes, $S - T_0$ mixing is driven by components of $\Delta B_{\text{nuc}}$ along the total field (applied plus Overhauser fields). In contrast, mixing of $S$ with $T_+$ (the $m_s = +1$ triplet), which occurs at a lower negative, field-dependent value of $\epsilon$ (red line in Fig. 1(b)) where Zeeman splitting matches exchange, is driven by components of $\Delta B_{\text{nuc}}$ transverse to the total field. Measuring the degree of evolution out of the prepared $S$ state following separation, by measuring the return probability to the (2,0) state, can be used to determine $\Delta B_{\text{nuc}}$.
charge configuration after a certain separation time, effectively measures these components of the Overhauser field difference in the two dots. Measurement is carried out by moving the system to position M in (2,0) for a time $\tau_M = 5 \mu s$, during which only S return to (2,0) with appreciable probability. The spin state—triplet or singlet—is thereby converted to a charge state—(1,1) or (2,0), respectively—which is detected by the rf-QPC.

Figures 1(c, d) show the time-averaged $V_{rf}$ as a function of gate voltages $V_L$ and $V_R$. Once calibrated, $V_{rf}$ gives the probability $1 - P_S$ that a prepared singlet evolved into a triplet during the separation time $\tau_S$. Inside the readout triangle (see Fig. 1(c)), triplet states remain blocked in (1,1) for a time $T_1 \gg \tau_M$ [14]. Similarly, inside the rectangular region indicated in Fig. 1(d), the prepared singlet mixes with $T_+$ and becomes blocked in (1,1). Calibration of $V_{rf}$ uses the signal in (2,0) outside the readout triangle, where fast, spin-independent relaxation occurs via (1,0) or (2,1), to define $P_S = 1$, and the region within (1,1) to define $P_S = 0$.

Fitting $P_S(\tau_S)$ averaged over tens of seconds with a gaussian [9] [13] (Fig. 1(e)) gives $T_1^* = \hbar/(g\mu_B B_{nuc}) \sim 15$ ns corresponding to $B_{nuc} \sim 1.6$ mT ($N \sim 6 \times 10^6$), where $g \sim -0.4$ is the electron g-factor and $\mu_B$ is the Bohr magneton. The effect of finite $T_1$ on the calibration of $P_S$ can be accounted by introducing a factor $C = (1 - e^{-\tau_M/T_1})/T_1/\tau_M$ [14] that relates $P_S$ to the value $P_S^* \sim T_1$, $1 - P_S \sim (1 - P_S^*)C$. The dependence of $P_S$ on $\tau_M$ (for a fixed $T_1 \sim 16 \mu s$ and $\tau_S = 50$ ns) is shown in Fig. 1(f). Applying this factor to Fig. 1(e) gives $P_S^*(\tau_S \gg T_1^*) = 1/3$, the expected value without normalizing the sensor output.

With less averaging, $P_S$ shows fluctuations that reflect fluctuations of Overhauser field components. Figure 2 shows a slice through the readout triangle, obtained by rastering $V_L$ at fixed $V_R$ with $B = 100$ mT, $\tau_S = 25$ ns. At $B = 100$ mT, fluctuations in $P_S$ have a flickering appearance with broadband time dependence extending to

![FIG. 1: (Color online) (a) Schematic energy diagram of the two-electron system. Inset: false-color SEM image of a double-dot with integrated rf-QPC charge sensor similar to the one measured (scale bar is 500 nm). (b) Gate-pulse cycle that is used to prepare (P) the (2,0) singlet, separate (S) into (1,1), either to the $S-T_0$ degeneracy (green dashed line) or the $S-T_+$ degeneracy (red dashed line), and return to (2,0) for measurement (M). (c) rf-QPC readout, $V_{rf}$, around the (1,1)-(2,0) transition during application of the cyclic gate-pulse sequence, showing the readout triangle indicated with white lines ($B = 0$ mT; $\tau_S = 50$ ns). A background plane has been subtracted. (d) $V_{rf}$ as in (c), but for S at the $S-T_+$ degeneracy ($B = 10$ mT). (e) Average value of $P_S(\tau_S)$ at $B = 0$, $\tau_M = 2 \mu s$. Red line is a fit to the theoretical gaussian form. (f) Average value of $P_S(\tau_M)$ showing contrast dependence, $\tau_S = 50$ ns. Red line is a fit to the exponential form (see main text).](image1)

![FIG. 2: (Color online) (a) rf-QPC sensor output $V_{rf}$ as a function of $V_L$ and $V_R$ with gate-pulse cycle applied ($\tau_S = 25$ ns, $\tau_M = 1.6 \mu s$, $B = 100$ mT). Color scale as in Fig. 1. (b) Repeated slices of $V_L$ with $V_R = -709$ mV as a function of time. Markers on left axis correspond to markers in (a). (c) Sensor output calibrated to $P_S$ (blue) along with a measurement of the background rf-QPC noise (pink) from (b) at arrow positions. (d) Similar to (b) but for $B = 0$, color scale same as in Fig. 1. (e) Similar to (b) but with S-point at $S-T_+$ degeneracy, $B = 100$ mT, color scale same as in Fig. 1.](image2)
FIG. 3: (Color online) (a) Power spectra of $P_S$ at various magnetic fields, $\tau_S = 25$ ns. Spectra obtained by FFT (with Hamming window) of average of 8 traces sampled at 10 kHz. Background measurement noise (BG) found by setting $\tau_S = 1$ ns at $B = 100$ mT. Inset: numerical simulation results for corresponding magnetic fields: $B = 0$ (pink), $B = 5$ mT (blue), $B = 10$ mT (green), $B = 100$ mT (red). (b) Autocorrelation $P_S$ for $\tau_S = 25$ ns and $B = 100$ mT (red curve). Model function (Eq. 1) (brown) and Monte Carlo result (black).

several seconds. Comparing the quieter (pink) trace in Fig. 2c, for point M such that $(1, 1)$ always returns to $(0, 2)$, to the fluctuating (blue) trace, where return to $(0, 2)$ requires $S - T_0$ mixing by Overhauser fields, we see that the amplitude of the fluctuating signal (blue) is $\sim 100$ times larger than the background noise of the charge sensor. At $B = 0$, slices across the readout triangle does not show a flickering (large, low-frequency) $P_S$ signal (Fig. 2(d)). Figure 2(e) shows slices across the $S - T_+\tau$ resonance (see Fig. 1(d)). Here also, $P_S$ also does not have a flickering appearance, independent of $B$, reflecting rapid fluctuations of transverse components of $\Delta B_{\text{nuc}}$. We avoid rapidly cycling through the $S - T_+$ transition, which can produce DNP [20].

To investigate the spectral content of $P_S$ fluctuations, fast Fourier transforms (FFTs) of $V_{\text{rf}}$ are taken with $V_L$ and $V_R$ positioned to sample the center of the readout triangle. Figure 3(a) shows power spectra of $P_S$, with $\tau_S = 25$ ns, over the range $B = 0$ - 100 mT. Measurement at $\tau_S = 1$ ns, where $P_S \sim 1$, has a 1/f form and is identical to the noise measured outside the readout triangle, and constitutes our background of instrumental noise. At $B = 0$ no spectral content above the 1/f background noise is seen (Fig. 2(a)). With increasing $B$, an increasing spectral content is observed below $\sim 100$ Hz. For $B > 20$ mT, the spectra become independent of $B$. The dependence of the power spectrum of $P_S$ on separation time $\tau_S$ is shown in Fig. 4. We found that the largest fluctuations over the greatest frequency range occur for $\tau_S \sim T_+^* \sim 15$ ns, and these fluctuations show a roughly 1/f$^2$ spectrum. Spectra were also obtained out to 100 kHz (not shown) where no additional high frequency components were observed above the background noise. For $\tau_S < T_+^*$, $P_S$ remains near unity with few fluctuations; For $\tau_S > T_+^*$ low-frequency content is suppressed while components in the range $1 - 10$ Hz are enhanced.

We model fluctuations in $P_S$ as arising from the dynamic Overhauser magnetic field in thermal equilibrium. A classical Langevin equation is used to describe fluctuations of $\Delta B_{\text{nuc}}$ arising from nuclear spin diffusion on distances much larger than the lattice spacing and times much longer than the time-scale set by nuclear dipole-dipole interaction. For $B \gg B_{\text{nuc}}$, correlations of the Overhauser field can be evaluated analytically in terms of a dimensionless operator $\hat{A}_z^2$ for each nuclear spin species $\beta$, where $\sum_{\beta} x^\beta \hat{A}_z^2 = B^z_{\text{nuc}}/B_{\text{nuc}}$ and similarly for the right dot, with $x^{71}\text{As} = 1, x^{75}\text{Ga} = 0.6, x^{71}\text{As} = 0.4$. This gives $\langle \hat{A}_z^2(t + \Delta t) \hat{A}_z^2(t) \rangle = \langle (1 + \Delta t/\Delta t_{\beta}) \rangle^{-1}$, at time difference $\Delta t$, where $\Delta t_{\beta}$ is the species-dependent spin diffusion coefficient, $\sigma_\perp$ is the electron wave function spatial extent perpendicular to the 2DEG (and along the external field) and $\sigma_\parallel$ is the wave function extent in the plane of the 2DEG, assumed symmetric in the plane. Brackets $\langle \ldots \rangle$ denote averaging over $t$ and nuclear ensembles.

Statistics of $P_S$ for $S - T_0$ mixing are found using the $z$-component of the Overhauser operators, $\Delta \hat{A}_z = \sum_{\beta} x^\beta (\hat{A}_z^2 - \hat{A}_z^2)$. For gaussian fluctuations and a species-independent diffusion constant, $D$, this gives a mean $\langle P_S \rangle = \frac{1}{2} [1 + e^{-2G^2(\Delta \hat{A}_z^2)}]$ and autocorrelation $\langle P_S(t + \Delta t) P_S(t) \rangle - \langle P_S \rangle^2$

$$= \frac{e^{-4G^2(\Delta \hat{A}_z^2)}}{4} \left[ \cosh(4G^2(\Delta \hat{A}_z(t + \Delta t) - \Delta \hat{A}_z(t))) - 1 \right],$$

where $G = \tau_S/T_+^*$ is a gain coefficient. The autocorrelation function at $B = 100$ mT shown in Fig. 3(b) is obtained by Fourier transforming the power spectrum [31]. We fit to the autocorrelation function using a contrast factor, $C$, (see Fig. 1(f) and discussion), and the diffusion coefficient, $D$, as fitting parameters. Wavefunction widths are taken from numerical simulations of the device [32], $\sigma_z = 7.5$ nm, $\sigma_\perp = 40$ nm. The fit gives $D \sim 10^{-13}$ cm$^2$/s, consistent with previous measurements on bulk GaAs samples using optical techniques [6].

In Eq. (1) the dependence on $\tau_S$ leads to a scaling of the correlation time of $P_S$ by $G^2$ to find the underlying Overhauser field correlation time. For fields $B > 20$ mT, the data in Fig. 3(b) indicate an autocorrelation time of $\sim 3$ s for $P_S$ corresponding to a time $\tau_S \sim 10$ s for $\Delta \hat{A}_z$ to decorrelate by half of its initial value.

Near $B \sim 0$, transverse components of the nuclear field lead to rapid dephasing of nuclear spins. In this regime, we use a Monte Carlo method to simulate nuclear dynamics [33]. Figure 3(b) shows that numerical and analytical
Clear spins and suppress long time correlations in $\Delta_{\text{nuc}}$ because nuclear fields at low $B$ can be understood as arising from the influence of the transverse spectral content as shown in the inset of Fig. 4. Similar behavior, though independent of $B$ and $\tau$, is observed in the spectra of $P_S$ at the $S_T$ resonance (not shown). Below $B \sim 10$ mT, an increased spectral content at frequencies between 1 - 10 Hz is observed in the experiment and theory. The frequency at which the spectra intersect however, remains constant ($\sim 1$ Hz) in the simulations but increases at low $B$ in the experimental data. We are able to approximate this behavior in the simulation by increasing the diffusion coefficient ($D \sim 10^{-12}$ cm$^2$/s at $B = 0$), implying an enhancement of diffusion, beyond typical values [6], as $B$ approaches zero. This may be due to the growing influence of non-secular terms in the dipole-dipole interaction at low magnetic field [8, 23]. Diffusion maybe further enhanced at low $B$ as a result of electron mediated flip-flop of nuclear spins [12, 34], an effect neglected in the simulation.

Finally, we model how the separation time for the two-electron spin state affects the power spectra. Simulated spectra are shown in the inset of Fig. 4 for $\tau_S = 1$ ns, 25 ns and 100 ns at $B = 100$ mT. Good agreement with experiment is achieved when again accounting for the additional $1/f$ noise and contrast reduction. We find that $\tau_S$ acts to filter fluctuations in $\Delta B_{\text{nuc}}$, so that for $\tau_S \gg T_2^*$, low frequency correlations in $\Delta B_{\text{nuc}}$ are suppressed in the spectra of $P_S$ (see Eq. 1). This filtering effect leads to the turn-over at $\sim 2$ Hz evident in the spectra for $\tau_S = 100$ ns. For $\tau_S \sim T_2^*$, little filtering occurs and the power spectra of $P_S$ reflect the underlying intrinsic fluctuations of the Overhauser magnetic field.

FIG. 4: (Color online) Power spectra of $P_S$ at $B = 100$ mT for separation times $\tau_S = 25$ ns (red) and $\tau_S = 100$ ns (blue). Setting $\tau_S = 1$ ns (black) yields background noise. Inset shows simulation results for $B = 100$ mT, $\tau_S = 25$ ns (red) and $\tau_S = 100$ ns (blue). Note the suppression of low-frequency content and enhancement of mid-frequency content for long $\tau_S$ in the experiment and simulation.

Experiment and theory show reduced low-frequency spectral content as $B$ decreases toward zero. This can be understood as arising from the influence of the transverse nuclear fields at low $B$, which rapidly dephase nuclear spins and suppress long time correlations in $\Delta B_{\text{nuc}}$. Similar behavior, though independent of $B$, is observed in the spectra of $P_S$ at the $S-T_\perp$ resonance (not shown).

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