Persistent narrowing of nuclear-spin fluctuations in InAs quantum dots using laser excitation

Bo Sun, Colin Ming Earn Chow, and Duncan G. Steel
The H. M. Randall Laboratory of Physics, The University of Michigan, Ann Arbor, Michigan 48109, USA

Allan S. Bracker and Daniel Gammon
Naval Research Laboratory, Washington D.C. 20375, USA

L. J. Sham
Department of Physics, The University of California, San Diego, La Jolla, California 92093, USA

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We demonstrate the suppression of nuclear spin fluctuations in an InAs quantum dot and measure the timescales of the spin narrowing effect. By initializing for tens of milliseconds with two continuous wave diode lasers, fluctuations of the nuclear spins are suppressed via the hole assisted dynamic nuclear polarization feedback mechanism. The fluctuation narrowed state persists in the dark (absent light illumination) for well over one second even in the presence of a varying electron charge and spin polarization. Enhancement of the electron spin coherence time ($T_{2}^*$) is directly measured using coherent dark state spectroscopy. By separating the calming of the nuclear spins in time from the spin qubit operations, this method is much simpler than the spin echo coherence recovery or dynamic decoupling schemes.

Single electrons trapped inside self assembled quantum dots form a well defined and optically accessible qubit, and are featured as the central element of many proposed quantum logic devices[1–7]. However, the electron spin in a quantum dot is coupled to many ($10^4 − 10^5$) nuclear spins, primarily via the Fermi contact hyperfine interaction, whose fluctuations form the main contribution to electron spin dephasing at cryogenic temperatures[8–11]. Thus, there has been considerable interest in suppressing electron spin dephasing by manipulating the nuclear spin ensemble[12–22], and recent results have shown that it is possible to protect the coherence of an ensemble of dots[23, 24] and even recover the electron spin coherence in a single quantum dot[25].

In this Letter, we use the hole assisted nuclear feedback mechanism[22, 26] to demonstrate the reproducible preparation of the nuclear magnetic field (Overhauser field) to a fluctuation suppressed state, considerably enhancing the electron spin coherence. This nuclear spin narrowing (NSN)[12–22] is accomplished without the creation of large nuclear spin polarizations. The spin narrowed state can be prepared in tens of milliseconds, and persists for well over a second even in the presence of a fluctuating electron charge and spin[27, 28]. We directly measure the enhanced $T_{2}^*$ using coherent dark state spectroscopy.

The sample is an InAs self assembled quantum dot embedded in a Schottky diode structure. A DC bias voltage applied across the sample charges the dot with a single electron and Stark shift modulation spectroscopy a large amplitude AC component (0.08VAC) at 3.5Khz directly measure the absorption spectrum[29]. Applying a 2.64T magnetic field in the Voigt profile (perpendicular to the growth axis) turns on spin flip Raman transitions between the spin ground states ($|X\pm\rangle$) and the charged exciton (trion) states ($|T\pm\rangle$) (Fig. 1a). We selectively excite a three-level lambda (Λ) subsystem (dashed outline in Fig. 1a) with narrow linewidth continuous wave lasers (Fig. 1b). The lasers are passed through acousto-optical shutters to individually gate the lasers on and off, decoupling NSN initialization from electron spin control and readout. Pump 1 is resonant with the H1 transition while pump 2 is slightly detuned from the V2 transition. The probe scans across the V2 transition. Figure 1c shows the gating of the lasers at each point of the absorption spectrum as the probe steps through the V2 resonance.

To measure the onset time of the spin narrowing effect, pump 1 and pump 2 are first gated on, populating the trion state. The trion’s unpaired hole interacts with the nuclei via a non-collinear hyperfine coupling, locking the Overhauser field to a value determined by the laser frequencies and produces spin narrowing via an intrinsic dynamic nuclear polarization feedback process[26]. Next, pump 2 is gated off and the probe is gated on for 25ms. When the pump and probe laser detunings are equal (at the two photon resonance), the electron spin forms a coherent superposition (dark state) which appears as a dip in the probe absorption spectrum[30, 31]. The strength of the dip is proportional to the electron spin decoherence rate ($1/T_{2}^*$)[1]. The strength of the dip is proportional to the electron spin decoherence rate ($1/T_{2}^*$)[1]. Since nuclear spin fluctuations contribute significantly to $T_{2}^*$, dark state spectroscopy is a sensitive measure of spin narrowing[12, 26]. Data is read through a lock in amplifier with the integration time constant set to a small value (5ms) to minimize the readout time and thus minimize any perturbations to the nuclei due to the readout. We only integrate the signal during the read out phase to maintain a large signal strength.
adequate evidence for this claim. Hence, the data in 2a are not
weaken the locking effect, requiring more time to gener-
tate the NSN state. However, there is an almost factor
of 7 decrease in the absolute signal strength, resulting in
between the dip and peak absorption reduces to
\[ \gamma \approx 700 \text{MHz} (150 \text{MHz}). \] Fitting the data to an exponential
curve) and pump 1 (pump 2) Rabi frequency (\( \Omega_R \))
and the probe are nearly resonant with transition \( V_2 \).
(c) Cartoon illustrating the laser illumination on the sample
at each point in the scan. During the Initialization stage, pump
1 and pump 2 produce a trion, whose hole component inter-
acts with the nuclear spins, preparing a NSN state. During
the Read-out stage, pump 1 and the probe then produce and
measure electron spin coherence, quantifying the narrowing of
the nuclear spin distribution. The nuclear spins (green arrows
in the background with large gaussian envelopes) start in a
state of large fluctuation. The NSN state is represented by
narrower gaussian envelopes, but maintains a similar average
field.

Figure 1 shows the laser gate timing diagram for each
point in a probe absorption spectrum.

Figure 2a shows, for various initialization times, the
measured probe absorption inside the dark state dip
normalized to the absorption at the Rabi sidebands at
sample temperatures of 5K (black curve) and 14K (red
curve) and pump 1 (pump 2) Rabi frequency (\( \Omega_R/2\pi \))
of 700MHz (150MHz). Fitting the data to an exponential
(solid lines), we extract an 100ms initialization time (at 5K).
We also fit the data to an exponential from which we can extract a
1/e time of 7 \( \pm 1 \)ms at 5K and 12 \( \pm 6 \)ms at 14K. The dashed
red line is a fit to the optical Bloch equations. (c) The black spectrum is taken using
a nominal scan range. The red spectrum has a reduced scan
range and shows a corresponding shift of the TPR, due to
probe effects on the Overhauser field. The blue arrows indicate
the location of pump 2.

Solving the optical Bloch equations\[32, 33\],
\[ i\hbar \frac{d\rho}{dt} = ([\hat{H}, \rho] + \text{decay}) \]
where \( \hat{H} \) is the Hamiltonian and \( \rho \) is the
density matrix, for a strong pump and weak probe in the
lambda system, we can find the probe absorption at the dark state dip (\( \alpha_{dip} \)) and the Rabi sideband (\( \alpha_{peak} \))\[20\]

\[
\alpha_{dip} = \alpha_0 \frac{\chi^2 \gamma_s + \gamma_t (\gamma_s^2)}{\chi^4 + 2\chi^2 \gamma_t \gamma_s + \gamma_t^2 \gamma_s^2} 
\]  
\[
\alpha_{peak} = \alpha_0 \frac{\chi^2 \gamma_s + \gamma_t (\gamma_s^2 + \chi^2)}{2\chi^2 \gamma_t \gamma_s + \gamma_t^2 \gamma_s^2 + (\gamma_t^2 + \gamma_s^2) \chi^2} 
\]

where \( \chi \) is half the pump 1 Rabi frequency, \( \gamma_t \) is the trion
dephasing rate, \( \gamma_s \) is the electron spin dephasing rate, and
\( \alpha_0 \) is a constant. In the limit where \( \gamma_s \ll \chi, \gamma_t \), the ratio
between the dip and peak absorption reduces to
\( \frac{\alpha_{dip}}{\alpha_{peak}} \approx \frac{\chi^4}{\gamma_t^2} \). Using this method, we estimate that \( \gamma_s/2\pi = 2\text{MHz} \)
for a 100ms initialization time (at 5K). We also fit the
dark state portion of the spectrum (solid red line in Fig.
2a) with the optical Bloch equations (the spectrum is too
distorted to fit directly) and find \( \gamma_s/2\pi = 6\text{MHz} \) with

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**Figure 1.** (Color online). (a) Four level energy diagram for
the trion system with a \( B=2.64\text{T} \) magnetic field applied in
the Voigt profile. The \( |T\pm> \) are the excited trion states
and the \( |X\pm> \) are the electron ground states. The relevant
temperature, we expect an increase in hole relaxation to
the nuclear spin distribution. The nuclear spins (green arrows
in the background with large gaussian envelopes) start in a
state of large fluctuation. The NSN state is represented by
narrower gaussian envelopes, but maintains a similar average
field.

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**Figure 2.** (Color online). (a) Measured dark state depths (relative
to the Rabi sidebands) as a function of the initialization
time at sample temperatures of 5K (black) and 14K (red).
The lines are fits to exponentials from which we can extract a
1/e time of 7 \( \pm 1 \)ms at 5K and 12 \( \pm 6 \)ms at 14K. The dashed
blue line indicates the relative depth of the dark state for
expected thermal value of the electron spin decoherence rate
of 360MHz. (b) Absorption spectra corresponding to select
points in (a). At short initialization times a second dip ap-
ppears to the blue (highlighted in blue), indicating bistability
of the Overhauser field. The red solid line is a fit to the op-
tical Bloch equations. (c) The black spectrum is taken using
a nominal scan range. The red spectrum has a reduced scan
range and shows a corresponding shift of the TPR, due to
probe effects on the Overhauser field. The blue arrows indicate
the location of pump 2.
an upper bound error of 14MHz. The expected thermal value of $\gamma_s/2\pi$ is 360MHz[26] at 5K and the relative dark state depth for this, calculated from simulations, is shown as the dashed blue line in Fig. 2a.

Figure 2b shows example spectra taken at 5K for several initialization times, where the blue arrow represents the position of pump 2 and each figure is an average of 40 scans. At short initialization times, there is a probe induced buildup of the Overhauser field which pushes the dark state to higher energy(highlighted in blue)[26]. At long initialization times, the probe effect is minimal. At intermediate initialization times, the appearance of the second dip may contribute to weakening of the central dip.

We note that while the dark state dip does not depend on the number of averages, which would be expected if there was an accumulated effect, we cannot discount memory effects in the dot which may impact the NSN dynamics measurement. The exponential function we use in Fig. 2b is only intended to give an indication of behavior and is not meant to represent a physical model.

Because the measured $1/e$ times are less than the read out time, the Overhauser field weakly locks to the probe laser as it steps through the absorption spectrum[26]. This locking only occurs over a limited range and results in the distorted lineshapes of Fig. 2b and c and shifts the TPR with a change in start position of the laser scan, seen in the red curve in Fig. 2 compared to the black curve. This does not affect the NSN measurement, only the average Overhauser field build up as there is no hole population at the TPR. Hence, the influence of the finite readout time does not impact our conclusion regarding the time scale of the preparation of the NSN state.

To measure the persistence of the NSN in the absence of laser interactions, we insert a dark period between the initialization and read out phases, indicated by the timing diagram in Fig. 3a. The black data in Fig. 3b is the spectrum with no dark period, and the red data is a comparison with no initialization or dark period. Using a pump 1 (pump 2) $\Omega_R/2\pi$ of 900MHz (650MHz) and an initialization time of 62.5ms, the absorption at both the dark state dip (green) and at the Rabi sidebands (blue) are plotted as a function of laser dark time in Fig. 3b. The average absorption of the Rabi sidebands (blue) is plotted along with the absorption in the dark state dip (green) as a function of the dark time. Clearly, NSN persists in the absence of laser illumination for well over 1s. The solid lines are an average. The black I is the error bar.

FIG. 3. (Color online). (a) We insert a dark period into the gating sequence to measure the persistence of the NSN as a function of laser dark time. (b) Black data is the absorption spectrum for 0ms dark time. The red data is a comparison where no initialization has occurred. Lines are guides to the eye. (c) The average absorption of the Rabi sidebands (blue) is plotted along with the absorption in the dark state dip (green) as a function of the dark time. Clearly, NSN persists in the absence of laser illumination for well over 1s. The solid lines are an average. The black I is the error bar.

to the electron charge and spin orientation[36]. The hole driven nonlinear hyperfine interaction leads to a reduction of nuclear spin fluctuations (NSN) without significantly modifying the mean Overhauser field, as seen by the position of the two photon resonance. In earlier observations of extended coherence times, such effects were driven by electron spin interactions and associated with changes in the nuclear spin polarization[20, 21].

In summary, we have shown that hole-assisted dynamical nuclear polarization feedback can be used to prepare the nuclear spins in a singly charged quantum dot into a spin narrowed state which can persist in the dark for over 1 second and has a preparation time of tens of milliseconds. The spin narrowing depends only on the hole spin and appears insensitive to the electron charge and spin orientation. This means that the NSN is potentially decoupled from quantum gate operations in which detuned pulsed lasers operate only on the TPR[2, 3]. Because these pulses specifically avoid populating the excited state and act primarily on the spin ground states, they should have minimal impact on the NSN. This approach can enhance the electron spin coherence prior to spin manipulation, thereby increasing the number of possible quantum computing operations without the need for spin echo coherence recovery or dynamic decoupling.
schemes.

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