Electrically detected spin echoes of donor nuclei in silicon

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The ability to electrically probe the spin properties of solid state systems underlies a wide variety of emerging technologies. Here, we demonstrate the electrical readout of the nuclear spin states of phosphorus donors in silicon in the coherent regime with modified Hahn echo sequences. We find that while the nuclear spins have electrically detected phase coherence times exceeding 2 ms, they are nonetheless limited by the artificially shortened lifetime of the probing donor electron.

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I. INTRODUCTION

Using spin to encode and process information lies at the heart of a range of emerging technologies.[1–5] While electron spins are an obvious choice for manipulating information, nuclear spins provide a robust system in which to store spin information for long periods of time.[6] However, while a number of optical[7,8] and quantum Hall based techniques[9] exist for reading the state of small ensembles of nuclear spins, until recently there has been no technique for doing so which is compatible with conventional electronic devices, particularly those in silicon. Using pulsed spin resonance, we recently demonstrated the ability to electrically measure the nuclear spin state of phosphorus donors in silicon[10] with a long spin lifetime. However, while the nuclear spin lifetime (T_1^n) imposes a limit on the storage of classical information, it is the nuclear spin phase coherence time (T_2^n) that sets a limit on the storage of quantum information, usually shorter than the spin lifetime. Here, we utilize a spin echo technique to show that electrical readout of donor nuclear spins is compatible with phase coherence times T_2^n exceeding 2 ms, nearly two orders of magnitude longer than that previously seen.[11] We also determine that the artificially shortened donor electron lifetime T_1^e limits T_2^n via hyperfine coupling to the nuclei,[6] and discuss ways to overcome this limitation.

To access the nuclear spin state of phosphorus donors in silicon, we exploit the precise hyperfine coupling between the donor nucleus and electron. By selectively exciting the donor electron dependent on the state of the nuclear spin, we obtain a sensitive probe of the nuclear spin.[10,12] A variety of methods can then be used to readout the selective excitation of the electron. For example, coupling the donor electron to the island of a nearby single electron transistor can allow single spin readout,[13] although readout of coherent states of the electron using this technique has not yet been reported in the literature. Hoehne *et al.*[11] have recently shown that electrical readout of coherent nuclear spin motion could be detected using spin-dependent recombination, although phase coherence times are limited to $T_2^n \sim 50\ \mu$s in those measurements.

Here, we utilize spin-dependent trapping of photoexcited electrons into the D^- state of the ^{31}P donor[14,15] [Fig. 1(a)]. We have previously used this mechanism to readout electron spin states with coherence times exceeding 150 μs,[14] and to determine the state of the nuclear spin following transfer and classical storage of the electron spin state.[10] The donors we measure are not restricted to the interface, and are therefore not influenced by near interface defects.[16–19] As described in detail in Ref. 14, the electron coherence time is in this case limited by the trapping of conduction electrons into the D^- state, a process which can be controlled by modifying the carrier density. The electron spin lifetime is determined by the occupation time of the electron. Figure 1(b) shows the result of an electrically detected inversion recovery experiment $[\pi(f_2^e) - \tau - \pi(f_2^e)]$ which allows us to determine the spin lifetime of the donor electrons; in these measurements $T_1^e = 1.86 \pm 0.02$ ms.

In our earlier work, we were unable to demonstrate coherent nuclear spin manipulation due to the spatial inhomogeneity of the rf radiation used to drive nuclear spin transitions. However, by utilizing spin echo techniques, we now show that electrically detected nuclear spin phase coherence times can exceed 2 ms.

II. ELECTRICALLY DETECTING NUCLEAR SPIN HAHN ECHOES

A phosphorus-doped silicon ($n[^{31}\mathrm{P}] = 10^{15}$ cm^{-3}) sample, described in detail in Ref. 10, was used for these experiments. Electrical contacts to the silicon are made with thin-film aluminum contacts. White light is shone onto the sample, and a constant current source with a slow time constant is used to provide a quasistatic photocurrent, $I = 200$ nA. Transient changes in this current, $\Delta I(t)$, are amplified and recorded with a digital oscilloscope. All experiments are performed at a temperature of 3.8 K. Electron spin transitions are driven using a custom-built spectrometer which operates at 240 GHz,[20,21] and which also contains an NMR coil to drive the nuclear spin transitions ($f_1^n = 88.60$ MHz, $f_2^n = 201.24$ MHz at $B = 8.56370$ T). The position and line shape was determined using the same procedure as in Ref. 10, and are almost identical to those measured in that work.

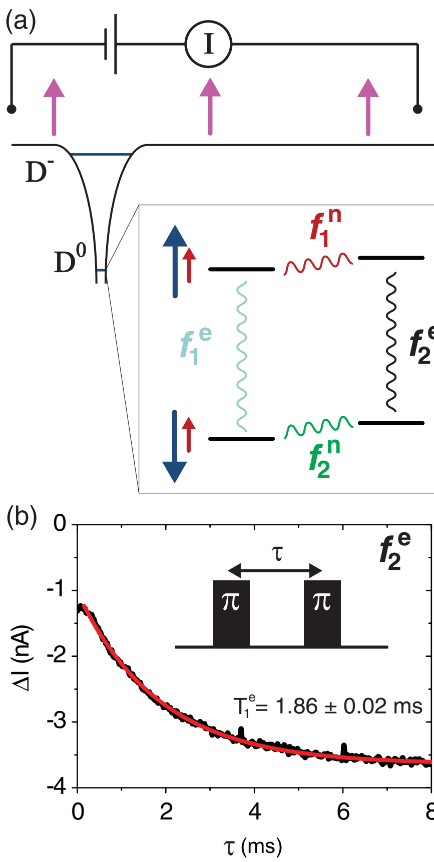

FIG. 1. (Color online) (a) Spin state of the P donor electron in silicon is detected by monitoring the photocurrent. Electrons from the conduction band are trapped in the D^- state only when they can form a singlet with the donor electron. The four eigenstates of the Si:P donor system are shown with large blue/dark gray arrows for the (spin 1/2) electron spin and smaller red arrows for the (spin 1/2) nuclear spin. The hyperfine interaction between the nucleus and electron lifts the degeneracy for the four transitions which can be driven using spin resonance. The nuclear spin state thus determines if resonance (and thus a change in the photocurrent) occurs at f_1^e or f_2^e. Probing one of these transitions therefore allows the nuclear spin state to be determined. For the experiments reported here, $f_2^e = 240$ GHz, $f_1^n = 88.60$ MHz, and $f_2^n = 201.24$ MHz. (b) An inversion-recovery experiment (f_2^e - τ - f_2^e) is used to find the lifetime of the electron spin $T_1^e = 1.86 \pm 0.02$ ms. The current change ΔI is the difference in the average current for the 200 μs before and after the final pulse.

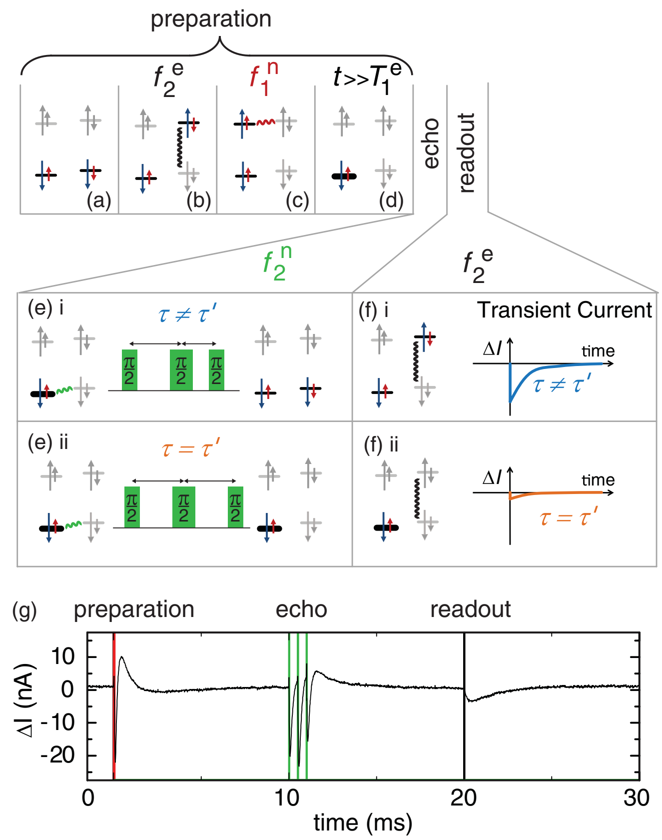

FIG. 2. (Color online) Pulse sequence for observing nuclear spin echoes. (a)–(d) Preparation of a polarized nuclear spin population is obtained by mapping the large electron spin polarization to the nuclear spins with a two-pulse sequence. (e) The nuclear spin echo is performed on the electron spin-down manifold. (f) Readout of the resulting nuclear spin population is achieved by selectively exciting the electron spins corresponding to nuclear spin down, and monitoring the current change through the sample after the excitation. (g) The change in current through the sample during an entire pulse sequence, with $\tau = 0.5$ ms. (Light gray spins represent nominally unpopulated states.)

To obtain a starting point for our measurements, we first polarize the nuclear spin [Figs. 2(a)–2(d)]. While this could be achieved in a number of ways,[22,23] we make use of the fact that the donor electrons are nearly entirely polarized by implementing a simple swap pulse sequence [$\pi(f_2^e)$–$\pi(f_1^n)$] (Ref. 24), similar to the sequence used in recent related work,[10,25,26] which results in an excess of nuclear spin up.

Following nuclear spin polarization, the system is allowed to recover for $t_{\rm wait} = 10$ ms, long enough for the electrons to relax to the ground state ($t_{\rm wait} > T_1^e$), but still much shorter than the nuclear spin lifetime ($t_{\rm wait} < T_1^n$) so that the polarization is maintained. A Hahn echo is then performed on the f_2^n transition, consisting of a $\pi/2 - \tau - \pi - \tau' - \pi/2$ pulse sequence.[27] A perfect echo should leave the nuclear spin polarization unchanged [Fig. 2(e)ii], whereas a nonoptimal echo sequence (e.g., with $\tau \neq \tau'$) should result in a decrease in the polarization, as some nuclear spin population returns to the spin-down state [Fig. 2(e)i]. To determine the resulting nuclear spin polarization, a readout pulse $\pi(f_2^e)$ is applied 10 ms after the start of the echo sequence. The magnitude of the change in current following the readout pulse is proportional to the nuclear spin-down population.

Figure 2(g) shows the current through the sample during one such polarization-echo-readout sequence. The transient behavior at 0 and 10 ms results from the application of the rf radiation used to drive nuclear spin transitions. It is nonresonant and most likely due to resistance changes from heating the sample. The current change at 20 ms is due to the electron trapping described above, and only occurs when the electron spin resonance conditions are matched (in this case for the f_2^e transition).

To observe the spin echo, the transient current ΔI following the readout pulse is measured[28] as a function of $\tau - \tau'$ for a fixed τ. Figure 3(a) shows ΔI during an echo sequence with

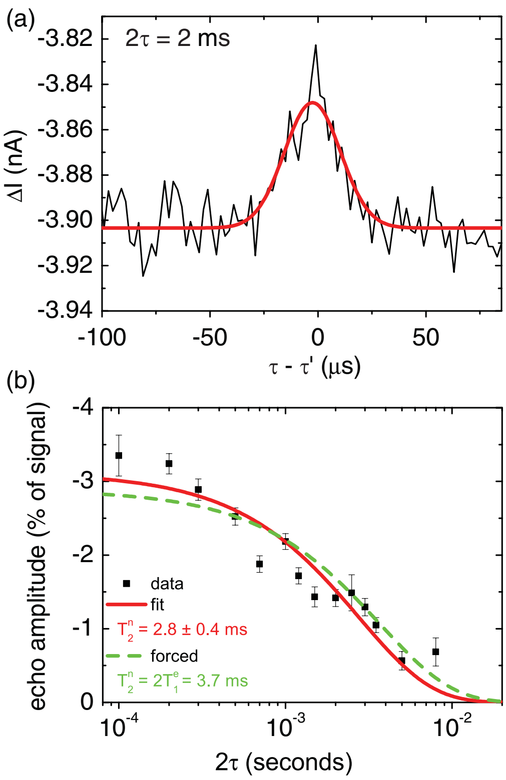

FIG. 3. (Color online) Nuclear echoes. (a) The change in photocurrent following the readout pulse for an echo sequence with $\tau = 1$ ms. A clear echo is seen when $\tau = \tau'$. (b) The amplitude of the echo is plotted for a range of τ. The data are fit with a simple exponentially decaying function, yielding a nuclear spin coherence time $T_2^n = 2.8 \pm 0.4$ ms (red solid line). The data are fit nearly as well if T_2^n is fixed to be $2T_1^e$ (green dashed line). Note the logarithmic scale on the x axis.

$\tau = 1$ ms. We expect rephasing to occur when $\tau - \tau' = 0$, and an echo is indeed observed when this condition is satisfied. The direction of the signal change (i.e., ΔI becomes smaller) is as expected. We note that the magnitude of the signal does not go to zero at the peak of the echo, and we attribute this to the nonideal pulses used to drive both electronic and nuclear spin transitions.

To determine the phase coherence of the nuclear spin system, the echo sequence was repeated for a range of τ. Figure 3(b) shows the echo amplitude $\Delta I(\tau = \tau') - \Delta I(\tau \neq \tau')$ as a percentage of $\Delta I(\tau \neq \tau')$, obtained by fitting each echo with a Gaussian peak function. The data are well fit by a simple exponential decay with a time constant $T_2^n = 2.8 \pm 0.4$ ms. This represents an increase of nearly two orders of magnitude for electrical readout of nuclear spin phase coherence of donors in silicon.

III. CHARGE CARRIER INDUCED DECOHERENCE

We note that, although long, the T_2^n we measure is nonetheless shorter than has been previously measured for donors in silicon using conventional means.[6] A number of processes exist which limit T_2^n in those measurements, but the fundamental limit seems to be the lifetime of the hyperfine coupled donor electron,[6,23] i.e., $T_2^n \leqslant 2T_1^e$. In the experiments reported here, the nuclear coherence time measured is indeed very close to twice the spin lifetime of the coupled donor electron, $T_2^n = 2.8 \pm 0.4$ ms $\approx 2T_1^e = 3.72 \pm 0.04$ ms [dashed line, Fig. 3(b)], indicating that this process is also the dominant dephasing mechanism in this work. The mechanism by which the nuclear spin is decohered by electron relaxation is described in Ref. 6, and we anticipate that increasing T_1^e should lead to longer T_2^n.

It is important to note that the electron spins in these experiments have artificially shortened lifetimes due to their interaction with the photoexcited conduction electrons required for readout. Figure 4 shows T_1^e measured by conventional pulsed electron spin resonance both in the dark and with the illumination required for electrical readout. In these experiments, the light intensity was optimized to maximize the reduction in T_1^e and resulted in a nearly two order of magnitude reduction at $T = 4$ K.

A number of modifications to the experiment described here would assist in obtaining these longer coherence times. A pulsed photoexcitation scheme could be utilized to enable readout only when required, retaining longer coherence times useful for computing and storage otherwise. Utilizing a MOSFET structure[29–31] would also allow this modification, as well as provide more accurate control of the free carrier density (and thus scattering time between free and donor electrons), which could be used to tune the nuclear spin lifetime.[32]

IV. SUMMARY

To summarize, we have demonstrated electrically measured spin echoes from phosphorus donor nuclear spins in silicon. We find a spin phase coherence time $T_2^n = 2.8 \pm 0.4$ ms. Evidence suggests that T_2^n is limited by the lifetime of the hyperfine coupled donor electron, which bodes well for increasing this time by utilizing methods to reduce the free electron density when readout is not required, for example, by utilizing pulsed photoexcitation or MOSFET style devices. Finally, we note that, as well as the relevance to technological devices based on spin, the techniques used here may find

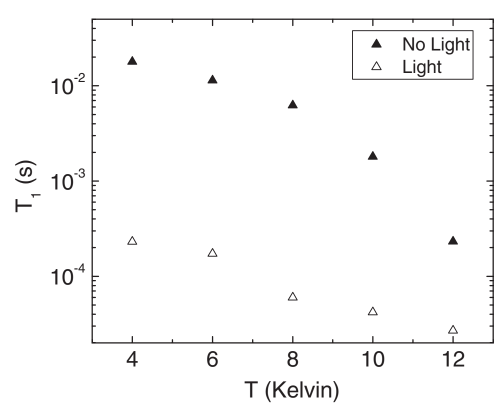

FIG. 4. Spin lifetime T_1^e of phosphorus donor electrons measured with an inversion recovery sequence at a resonance frequency of 240 GHz with conventional pulsed electron spin resonance. The measurements are performed both in the dark (closed data points) and while shining light onto the sample (open data points).

applications as tools for investigating the physics of systems comprised of a small number of spins, where conventional spin resonance techniques are not suitable.

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[1]S. A. Wolf, D. D. Awschalom, R. A. Buhrman, J. M. Daughton, S. von Molnr, M. L. Roukes, A. Y. Chtchelkanova, and D. M. Treger, Science **294**, 1488 (2001).

[2]I. Žutić, J. Fabian, and S. Das Sarma, Rev. Mod. Phys. **76**, 323 (2004).

[3]B. E. Kane, Nature **393**, 133 (1998).

[4]V. A. Dediu, L. E. Hueso, I. Bergenti, and C. Taliani, Nat. Mater. **8**, 707 (2009).

[5]T. Ladd, F. Jelezko, R. Laflamme, Y. Nakamura, C. Monroe, and J. O'Brien, Nature **464**, 45 (2010).

[6]J. J. L. Morton, A. M. Tyryshkin, R. M. Brown, S. Shankar, B. W. Lovett, A. Ardavan, T. Schenkel, E. E. Haller, J. W. Ager, and S. A. Lyon, Nature **455**, 1085 (2008).

[7]P. Neumann, J. Beck, M. Steiner, F. Rempp, H. Fedder, P. R. Hemmer, J. Wrachtrup, and F. Jelezko, Science **329**, 542 (2010).

[8]M. Steger, T. Sekiguchi, A. Yang, K. Saeedi, M. E. Hayden, M. L. W. Thewalt, K. M. Itoh, H. Riemann, N. V. Abrosimov, P. Becker, and H.-J. Pohl, J. Appl. Phys. **109**, 102411 (2011).

[9]G. Yusa, K. Muraki, K. Takashina, K. Hashimoto, and Y. Hirayama, Nature **434**, 1001 (2005).

[10]D. R. McCamey, J. van Tol, G. W. Morley, and C. Boehme, Science **330**, 1652 (2010).

[11]F. Hoehne, L. Dreher, H. Huebl, M. Stutzmann, and M. S. Brandt, Phys. Rev. Lett. **106**, 187601 (2011).

[12]M. Sarovar, K. C. Young, T. Schenkel, and K. B. Whaley, Phys. Rev. B **78**, 245302 (2008).

[13]A. Morello, J. J. Pla, F. A. Zwanenburg, K. W. Chan, K. Y. Tan, H. Huebl, M. Möttönen, C. D. Nugroho, C. Yang, J. A. van Donkelaar, A. D. C. Alves, D. N. Jamieson, C. C. Escott, L. C. L. Hollenberg, R. G. Clark, and A. S. Dzurak, Nature **467**, 687 (2010).

[14]G. W. Morley, D. R. McCamey, H. A. Seipel, L. C. Brunel, J. van Tol, and C. Boehme, Phys. Rev. Lett. **101**, 207602 (2008).

[15]D. D. Thornton and A. Honig, Phys. Rev. Lett. **30**, 909 (1973).

[16]T. Schenkel, A. M. Tyryshkin, R. de Sousa, K. B. Whaley, J. Bokor, J. A. Liddle, A. Persaud, J. Shangkuan, I. Chakarov, and S. A. Lyon, Appl. Phys. Lett. **88**, 112101 (2005).

[17]R. de Sousa, Phys. Rev. B **76**, 245306 (2007).

[18]S.-Y. Paik, S.-Y. Lee, W. J. Baker, D. R. McCamey, and C. Boehme, Phys. Rev. B **81**, 075214 (2010).

[19]H. Huebl, F. Hoehne, B. Grolik, A. R. Stegner, M. Stutzmann, and M. S. Brandt, Phys. Rev. Lett. **100**, 177602 (2008).

[20]J. van Tol, L.-C. Brunel, and R. J. Wylde, Rev. Sci. Instrum. **76**, 074101 (2005).

[21]G. W. Morley, L.-C. Brunel, and J. van Tol, Rev. Sci. Instrum. **79**, 064703 (2008).

[22]D. R. McCamey, J. van Tol, G. W. Morley, and C. Boehme, Phys. Rev. Lett. **102**, 027601 (2009).

[23]J. van Tol, G. W. Morley, S. Takahashi, D. R. McCamey, C. Boehme, and M. E. Zvanut, Appl. Magn. Reson. **36**, 259 (2009).

[24]E. R. Davies, Phys. Lett. A **47**, 1 (1974).

[25]S. Simmons, R. M. Brown, H. Riemann, N. V. Abrosimov, P. Becker, H.-J. Pohl, M. L. W. Thewalt, K. M. Itoh, and J. J. L. Morton, Nature **470**, 69 (2011).

[26]G. W. Morley, J. van Tol, A. Ardavan, K. Porfyrakis, J. Zhang, and G. A. D. Briggs, Phys. Rev. Lett. **98**, 220501 (2007).

[27]The pulse times used in this work (in μs) are $\pi(f_2^e) = 1.2$, $\pi(f_2^n) = 25$, $\pi/2(f_2^n) = 15$, and $\pi(f_1^n) = 50$. Approximately 10% of donor nuclear spins are rotated following a $\pi/2$ pulse.

[28]We average the change in current for 300 μs following the readout pulse. Each point averaged 20 times, with a repetition rate of 30 ms.

[29]C. C. Lo, J. Bokor, T. Schenkel, A. M. Tyryshkin, and S. A. Lyon, Appl. Phys. Lett. **91**, 242106 (2007).

[30]L. H. Willems van Beveren, H. Huebl, D. R. McCamey, T. Duty, A. J. Ferguson, R. G. Clark, and M. S. Brandt, Appl. Phys. Lett. **93**, 072102 (2008).

[31]C. C. Lo, V. Lang, R. E. George, J. J. L. Morton, A. M. Tyryshkin, S. A. Lyon, J. Bokor, and T. Schenkel, Phys. Rev. Lett. **106**, 207601 (2011).

[32]N. Stemeroff and R. de Sousa, Phys. Rev. Lett. **107**, 197602 (2011).