Electrical detection of spin echoes for phosphorus donors in silicon

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Abstract

The electrical detection of spin echoes via echo tomography is used to observe decoherence processes associated with the electrical readout of the spin state of phosphorus donor electrons in silicon near a SiO₂ interface. Using the Carr-Purcell pulse sequence, an echo decay with a time constant of 1.7 ± 0.2 µs is observed, in good agreement with theoretical modeling of the interaction between donors and paramagnetic interface states. Electrical spin echo tomography thus can be used to study the spin dynamics in realistic spin qubit devices for quantum information processing.

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A fundamental requirement for physical realizations of quantum computers is that the decoherence time of the qubits significantly exceeds typical gate operation times. As an example, decoherence times of the electron spin of phosphorus donors of up to 60 ms have been reported in bulk isotopically pure $^{28}$Si crystals [1]. Since typical $\pi$ pulse lengths used to manipulate such spins via magnetic resonance are of the order of 100 ns, donor spin states clearly satisfy the above requirement. The effects of spin-spin interactions between neighboring P donors as well as with the nuclear magnetic moments of the $^{29}$Si isotope, which is present with an abundance of 4.6% in natural silicon, on the decoherence have already been investigated in detail [2, 3]. However, the existing bulk measurements are not directly relevant for quantum computing applications, since in more realistic qubit devices such as proposed by Kane [4], the donors have to be in close proximity to a Si/SiO$_2$ interface, which is required for the electrical isolation of gate electrodes used to address the donor qubits [4, 5]. Therefore, a coupling to paramagnetic interface states such as the P$_{b0}$ centers [6, 7] will provide an additional source of decoherence [8, 9]. Stegner et al. [10] have actually used this donor-interface state interaction for a purely electrical readout of the donor spin states by pulsed electrically detected magnetic resonance (pEDMR), which has the potential to overcome the sensitivity limitations of conventional electron spin resonance (ESR) down to the single spin detection limit [11]. In this paper, we demonstrate that pulsed EDMR experiments can also be employed to investigate decoherence processes using a Carr-Purcell pulse sequence in combination with echo tomography [12]. In Si/SiO$_2$ heterostructures with a natural isotope composition, where the P donors are located within 15 nm of the interface, we find that the Carr-Purcell echo at 6.5 K decays with a time constant of $1.7 \pm 0.2 \mu$s and quantitatively discuss this result taking into account decoherence and recombination processes. In the framework of quantum information processing, this result shows that the spin read-out via P-P$_{b0}$ spin pairs could be applied at least to fundamental algorithms requiring up to $30\pi$ pulses. More importantly, this work shows that with the help of advanced pulse sequences combined with echo tomography valuable new information on the complex charge carrier and spin dynamics can be obtained by EDMR.

To detect the donor spin information electrically, we use a spin-to-charge conversion method based on the Pauli exclusion principle. The P donor and a nearby P$_{b0}$ center form a correlated spin pair as shown in Fig. 1(a). Since a doubly occupied, negatively charged P$_{b0}^-$ state as the final state of the spin-to-charge conversion has a total spin $S = 0$, the transition
of the electron from a P donor to a neutral P$_{b0}$ center is forbidden for triplet configurations of the P-P$_{b0}$ pairs, while for singlet P-P$_{b0}$ pairs the transition can occur, thus influencing the electronic transport properties of the sample [13]. Under spin resonance conditions the relative orientation in the spin pair can be changed coherently e.g. from the triplet to the singlet configuration by a $\pi$-pulse, resulting in a change of the rate of the spin-dependent transition sketched in Fig. 1 a), which in turn is observable as a resonant change of the conductivity [10].

Decoherence is usually studied with the help of a Carr-Purcell echo experiment with a $\pi/2$-$\tau_1$-$\pi$-$\tau_2$-echo pulse sequence, where $\pi/2$ and $\pi$ denote the rotation angle of the spin system induced by resonant microwave pulses and $\tau_1$ and $\tau_2$ are the free evolution periods between the pulses [14]. Such a Carr-Purcell echo is shown in Fig. 1 b) for an ensemble of identical spins (e.g. the spins of phosphorus donors) plotted in a Bloch sphere, starting e.g. with the spin ensemble in the ”down” eigenstate $\left|\downarrow\right>$ [25]. The microwave pulses are assumed to rotate the spins around the $x$-axis of the Bloch sphere. The echo develops in the $x$-$y$ plane of the Bloch sphere, giving rise to a pulse in the transverse magnetization or coherence, which is easily detectable in conventional ESR. However, the spin-to-charge conversion process employed here is sensitive to the singlet/triplet symmetry of a spin pair, which is not influenced by the orientation in the $x$-$y$ plane of the Bloch sphere of one constituent spin if the other spin is left in an eigenstate. A successful detection of such echoes via charge transport therefore requires so-called echo tomography [12], where after the second free evolution period $\tau_2$ a final $\pi/2$-pulse rotates the spin system back into singlet or triplet eigenstates of the pair. Again for an ensemble of spins such as the P donor spins the effect of a final $\pi/2$ pulse is shown in Fig. 1 c) on a Bloch sphere in more detail. For $\tau_2 \ll \tau_1$ and $\tau_2 \gg \tau_1$, no echo has developed in the $x$-$y$ plane so that after the final $\pi/2$ pulse, the spins of the ensemble point to all directions in the $x$-$z$ plane of the Bloch sphere. Both, triplet and singlet configurations will therefore be found in ensembles of P-P$_{b0}$ spin pairs. However, for $\tau_2 = \tau_1$, an echo has developed, so that after the final $\pi/2$ pulse, the spin ensemble is in the original $\left|\downarrow\right>$ eigenstate again. If the P$_{b0}$ partner in the spin state is also in the $\left|\downarrow\right>$ state, we find only the triplet configuration $\left|\downarrow\downarrow\right>$ for the P-P$_{b0}$ spin pairs. A comparison of the two cases with $\tau_2 = \tau_1$ and $\tau_2 \neq \tau_1$ shows that via the application of the final $\pi/2$ pulse, echoes can be formed also in the singlet/triplet symmetry of spin pairs and therefore are accessible to purely electrical detection.
In electrically detected magnetic resonance, until now only rotary spin echoes have been reported \[15\], where the spin system is continuously driven by the microwave and the echo develops when the microwave-induced rotation is canceled out by an equally long reversed rotation realized by a 180° phase shift in the microwave. While these echoes in principle are also able to quantitatively measure decoherence \[16\], the use of pulse sequences including free evolution periods together with echo tomography is more versatile and in principle allows other properties such as the exchange interaction in the P-P\(_{b0}\) spin pair or the superhyperfine interaction with \(^{29}\)Si to be investigated by double electron electron resonance (DEER) \[17\] and electron spin echo envelope modulation (ESEEM) \[3\] pulse techniques, respectively.

Due to RC timeconstants of the experimental system consisting of the sample and the measurement electronics, Carr-Purcell echoes cannot be observed in real time. Rather, current transients after the application of the microwave pulse sequence are measured, where the excited spin system relaxes back into the steady-state. Theory shows that the amplitudes of the multi-exponential decay are proportional to the deviation of the singlet/triplet configuration of the spin-system at the end of the pulse sequence from the steady-state configuration \[18\]. Therefore, measurements on long time scales compatible with the detection system allow to monitor the coherent manipulation of the spin system and its decoherence taking place on much shorter timescales.

To increase the signal-to-noise ratio, the current transients are integrated in a box-car type of analysis, yielding a characteristic charge \(Q\) as the primary result of the experiment. As shown in Fig. 1 d) for the coherent manipulation of the P spin by a microwave pulse of varying time \(\tau_{\text{Rabi}}\), \(Q\) is small if the manipulation brings the spin system back into the steady state. In contrast, the current transient and therefore \(Q\) is large when the spin system has been brought out of equilibrium by the microwave pulse. These electrically detected Rabi oscillations have been discussed in detail by Stegner et al. \[10\]. Due to the Zeeman interaction, the spin state \(|\downarrow\downarrow\rangle\) is the energetically lowest state of the P-P\(_{b0}\) spin pair and is therefore indicated as the steady-state configuration in Fig. 1 d).

We are now in the position to predict the experimental signature of the echoes discussed above. The pulse sequence \(\pi/2-\tau_1-\pi-\tau_2-\pi/2\) contains microwave pulses with a total length of \(2\pi\). Ideally, we therefore expect a value of \(Q\) after a Carr-Purcell echo sequence with \(\tau_1 = \tau_2\) equal to the \(Q\) found after a rotation by \(2\pi\) in a Rabi-flop experiment as shown in Fig. 1 e). For \(\tau_2 \gg \tau_1\) and \(\tau_2 \ll \tau_1\), our discussion of Fig. 1 c) above has shown that the spin pairs
are not in a steady-state configuration, and therefore a larger $Q$ is expected as the result of such Carr-Purcell sequences, as also sketched in Fig. 1(e).

The sample investigated here was grown by chemical vapor deposition and consists of a 15 nm thick silicon layer doped with phosphorus at $[P] = 10^{17}$ cm$^{-3}$ covered with a native oxide on top of a 500 nm thick nominally intrinsic buffer on a Si:B wafer (30 Ωcm) [19]. The measurements are performed at 9.765 GHz in a dielectric microwave resonator (Bruker ER 4118XMD5W1) between 5 K and 15 K under illumination with the broad optical spectrum of a tungsten lamp. The microwaves are generated by a HP83640A synthesizer and amplified by a traveling wavetube amplifier. For the conductivity measurements interdigit Cr/Au contacts with a periodicity of 20 µm covering an active device area of $2 \times 2.25$ mm$^2$ are used, so that $\approx 10^{10}$ P spins are probed. During the experiment, the sample is biased with 22 mV resulting in a current of $\approx 50$ µA, which is measured by a current amplifier. To record the current transients the output of the amplifier (500 kHz bandwidth) is connected via a passive Butterworth high-pass filter ($7^{th}$ order, $f_{3dB} = 400$ Hz) and a video amplifier (20 MHz bandwidth) to a digital storage oscilloscope. This allows to detect photocurrent transients after the microwave pulse, which under these conditions consist of a fast (characteristic time constant $t = 3$ µs) rise of the conductivity, a slower ($t = 11$ µs) fall overshooting the steady-state value followed by a very slow ($t = 140$ µs) rise back to the steady-state conductivity. An unambiguous assignment of the experimental time constants to singlet and triplet recombination times is not possible, in particular due to the limited bandwidth of the current amplifier prohibiting the observation of transients faster that 2-3 µs. While a full understanding of the time constants is certainly interesting in the long run, it is not required for the experimental detection of decoherence and the determination of the relevant time scales performed here. To obtain a sufficient signal-to-noise level the transients were accumulated with a repetition time of 350 µs. Control experiments at slower repetition rates showed that the results obtained were not influenced by this repetition time. The box-car type integration was performed over the positive part of the overall photocurrent transient from 2-22 µs. Rabi oscillations were used to determine the $\pi/2$-pulse time to 31 ns at the specific microwave power employed corresponding to a of the microwave $B_1$ field of 0.3 mT.

Figure 2 shows the Carr-Purcell echo tomography data $\Delta Q$ obtained after a substraction of a constant background from the Si:P/SiO$_2$ heterostructure. A first evolution period $\tau_1 = 200$ ns was selected and $\tau_2$ as well as the magnetic field $B_0$ were varied. As expected,
a single echo with $\Delta Q < 0$ is found for $\tau_1 = \tau_2 = 200$ ns for the 4.2 mT hyperfine split P resonances with the central $g$-factor $g = 1.9985$ at $B_0 = 351.2$ mT and $B_0 = 347.0$ mT and a broad $P_{10}$ resonance. In fact, two resonance lines at $g = 2.008$ ($B_0 = 347.4$ mT) and $g = 2.004$ ($B_0 = 348.1$ mT) are expected for the magnetic field oriented along the [110] axis of the Si sample [8]. Due to the high excitation power used for the pulses and their large inhomogeneous linewidth these two resonances are not resolved in Fig. 2, but rather appear as a single feature at $B_0 = 347.9$ mT. The full-width half-maximum temporal extent $\Delta \tau_{1/2}$ of the echo is correlated with the inhomogeneous Gaussian linewidth $\Delta B_{1/2}$ via $\Delta \tau_{1/2} = (2h)/(g\mu_B\Delta B_{1/2})$, where $g$ is the $g$-factor, $\mu_B$ is the Bohr magneton and $h$ is Planck’s constant [20]. For an inhomogeneous linewidth of 0.4 mT for the high-field P resonance at $B_0 = 351.2$ mT, a $\Delta \tau_{1/2}$ of 170 ns is expected, in good agreement with the value of 130 ns determined from the experimental echo data $Q$ for this resonance shown in more detail in Fig. 1 e) [26]. A quantitative comparison of $Q$ observed during the echo (Fig. 1 e) to the $Q$ observed in the Rabi oscillation (Fig. 1 d) demonstrates that the echo amplitude $\Delta Q$ is indeed as large as expected from the discussion above. However, the absolute values of $Q$ are higher in the echo experiment and correspond to $Q$ found in the Rabi oscillations at high $\tau_{Rabi}$. This is most probably caused by the limited attenuation of the microwave switch in the “off” state, which leads to a weak but continuous disturbance of the spin system also during the free evolution periods.

To determine the echo decay time in this system, the Carr-Purcell echo sequence is measured as a function of $\tau_1$. In all cases, the echo is observed at $\tau_1 = \tau_2$, however the echo intensity decreases for longer $\tau_1, \tau_2$. Figure 3 shows $\Delta Q$ as a function of the total free evolution period $\tau_1 + \tau_2$ again for the high-field P resonance at $B_0 = 351.2$ mT obtained after substraction of a linear background shown as a dashed line in Fig. 1 e). The decay of the echo amplitude is described well with a mono-exponential decay using a time constant $\tau_{echo} = 1.7 \pm 0.2 \mu s$ (solid line in Fig. 3). The same time constant is observed on the $P_{10}$ echo. The transients as well as the echoes and their decay are independent of temperature up to 12 K. Above 12 K, no echo is observed due to the decrease in the amplitudes of the underlying current transient.

At least two physical processes can contribute to the decay of the echo, namely (i) the actual decoherence usually denoted by the transverse relaxation time $T_2$ and (ii) the loss of spin-pairs through recombination. If the two processes are independent, Matthiessen’s rule
predicts that the faster process will govern the overall echo decay.

Let us first assume that the echo decay of caused by fast $T_2$ processes. Comparing the observed echo decay time $\tau_{echo}$ with reports on decoherence in Si:P from the literature, it becomes clear that the value reported here is significantly shorter than in earlier experiments via conventional pulsed ESR to values between 100 $\mu$s and 60 ms [1, 21]. Those experiments are typically performed at low doping concentrations ($< 2 \times 10^{16}$ cm$^{-3}$) and in bulk samples to ensure the isolation of the phosphorus spins. Chiba and Hirai [2] studied the decoherence time as function of the phosphorus doping concentration at 1.5 K and observed a significant decrease of $T_2$ by two orders of magnitude from 300 $\mu$s to 5 $\mu$s at a doping concentration of $1 - 2 \times 10^{17}$ cm$^{-3}$ similar to the doping concentration used in this experiment. Therefore, the $\tau_{echo}$ of $1.7 \pm 0.2$ $\mu$s determined here could already be accounted for by the high doping concentration.

Additionally, in this experiment the P donors are particularly close to the Si/SiO$_2$ interface expected to be necessary for the tuning of the hyperfine interaction and of the exchange between neighboring donors by gate potentials when using P in devices for quantum logic [4, 5]. De Sousa [9] has calculated the influence of magnetic noise caused by paramagnetic Si/SiO$_2$-interface states on the decoherence times of donors and estimates $T_2 = 4 \times 10^{-8}$ s $\times (d/\text{nm})^2$ for an interface spin density of $10^{11}$ cm$^{-2}$, where $d$ denotes the distance of the donors to the interface. For these interface state densities, Schenkel et al. [8] have determined the decoherence of Sb donors in $^{28}$Si by conventional pulsed ESR for Sb implanted to comparatively large depths of 50 and 150 nm, in good agreement with the theory. Taking into account the considerably smaller distance to the interface studied here and the slightly larger P$_{b0}$ density of $10^{12}$ cm$^{-2}$ of native oxides [22] our data can similarly be accounted for well by de Sousa’s prediction [9]. Furthermore, the temperature independence of $\tau_{echo}$ suggests that spin flip-flop processes between the P and the P$_{b0}$ spins.

Alternatively, the echo decay could also be due to recombination. Rate limiting for Carr-Purcell echoes would be the singlet recombination rate $r_S$ [18]. Since the singlet content of the spin pair during the evolution periods of our echo experiment is 1/4, $\tau_{echo} = 4/r_S$ in the case of slow $T_2$ processes. For P-P$_{b0}$ spin pairs, the singlet recombination time has not been determined independently. However, effective lifetimes of photo-generated charge carriers can be as short as 1 $\mu$s, as has been studied for P-doped emitters in crystalline Si solar cells [23], structures very similar to our test device. The value of $\tau_{echo}$ found would in this case
correspond to \( r_S = 2.3 \times 10^6 \) \(1/\text{s}\), a rate significantly higher than observable directly from the current transients. Since EDMR experiments on the identical structures using the inversion recovery pulse sequence are limited by the same \( r_S \) [24], we conclude that, at the densities of P and \( \text{P}_b\text{O} \) in our sample, indeed recombination determines the echo decay. Furthermore, the inversion recovery EDMR results suggest that microwave phase decay noise can be excluded as the origin of the echo decay.

As discussed, the P-\( \text{P}_b\text{O} \) pair interaction allows the purely electrical detection of the spin states of donor qubits. Irrespective of the actual microscopic mechanism, the echo decay time determined by pulsed EDMR is the relevant timescale on which such pairs can be used for quantum information processing. The \( \tau_{\text{echo}} \) found here allows to apply around 30 \( \pi \)-pulses of 60 ns length. While this is below the standard DiVincenzo requirement, this should be sufficient to demonstrate the basic useability of P donors in quantum information processing. Still, a variation of the phosphorus density, the density of the interface states, the isotopic composition of the host lattice and the illumination intensity should allow to reduce the decoherence processes and increase the recombination times in such structures extending the time scale on which P-\( \text{P}_b\text{O} \) pairs can be used for quantum logic.

In conclusion, we have demonstrated the electrical detection of Carr-Purcell echoes in 15 nm thin Si:P structures. The decay of the echo amplitude is mono-exponential with a decay time constant of \( 1.7 \pm 0.2 \) \(\mu\text{s}\). Several mechanisms such as the relatively high phosphorus doping concentration, the small P/SiO\(_2\)-interface distance, and the illumination of the sample will contribute to the overall decoherence process. The systematic determination of the echo decay time as a function of spin density, interface distance and illumination will provide further experimental data on the dynamics of the P-\( \text{P}_b\text{O} \) spin pairs, which are highly relevant for the use of this system for the read-out of donor spins. However, the results also directly address other donor-based quantum computer architectures with different spin read-out techniques, since \( \text{P}_b\text{O} \) paramagnetic centers are present at all Si/SiO\(_2\) interfaces. Furthermore, the determination of decoherence via Carr-Purcell echoes will allow a more detailed understanding of the complex charge carrier and spin dynamics in pulsed EDMR. Finally, the combination of echo tomography and pulsed electrically detected magnetic resonance opens the possibility to apply further pulse sequences including free evolution times to study spin-spin interactions in Si with unprecedented sensitivity.

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[25] Since only the relative orientation of the spin pairs is detected in EDMR, our arguments also apply to spins in the "up" state $|\uparrow\rangle$ and therefore no polarization of the spin ensemble is needed to observe echoes with this technique.

[26] The larger linewidth of the $P_{10}$ resonances leads to a narrower temporal extent of the echoes observed on these resonances as expected.
Figures

a) Schematic of the experimental setup with energy levels and labels.

b) Diagram showing the temporal evolution of the system with time labels and phases.

d) Graph showing the dependence of a parameter $Q$ on Rabi time $\tau_{Rabi}$.

e) Graph comparing $Q$ to $\tau_2$ with a highlighted region $\Delta Q$. 

c) Sequence of states before and after a final $\pi/2$ pulse.
FIG. 1: a) Spin-dependent transition involving the electron spin of a $^{31}$P donor and a paramagnetic state $P_{b0}$ at the interface between the crystalline Si (c-Si) and SiO$_2$. Since the final state is a doubly occupied diamagnetic $P_{b0}^-$, the transition can only occur for initial $P-P_{b0}$ spin pairs in a singlet configuration. Electron spin resonance of either $^{31}$P or $P_{b0}$ influences the spin configuration, and results in a resonant change of the transition rate which can be monitored by charge transport through the test structure. b) Carr-Purcell echo for an ensemble of identical spins plotted on a Bloch sphere. Before the second $\pi/2$ pulse, an echo develops in the $x$-$y$-plane, which is rotated to an eigenstate by the final "echo tomography" pulse, necessary for the electrical detection of the echo. The effect of a change in the second evolution time $\tau_2$ with respect to $\tau_1$ on the spin ensemble before and after the final $\pi/2$ pulse is shown in more detail in c). d) Charge $Q$ obtained from the integration of the current transients after microwave pulses resonant with the $^{31}$P donor spin ensemble (Rabi oscillations). For spin configurations of the $P-P_{b0}$ spin pair identical to the initial configuration (corresponding to $2\pi$, $4\pi$, ... pulses), the transients and therefore $Q$ are small. When the microwave pulse has brought the spin system out of equilibrium, $Q$ is large ($\pi$, $3\pi$, ... pulses). e) The total pulse length in a Carr-Purcell echo experiment including the tomography pulse is $2\pi$. If $\tau_2 = \tau_1$, the initial spin configuration of the $P-P_{b0}$ spin pair will be restored, while for $\tau_2 \neq \tau_1$, the spin system is out of equilibrium. A comparison with the Rabi oscillation in d) predicts a decrease of $Q$ under echo conditions $\tau_2 \approx \tau_1$. Parts d) and e) show the Rabi oscillation and the Carr-Purcell echo, respectively, as detected on the high-field resonance of the $^{31}$P donor electron spin. The amplitude $\Delta Q$ of the echo agrees well with the prediction.
FIG. 2: Electrically detected Carr-Purcell echoes for a first free evolution period of $\tau_1 = 200$ ns. A clear signature in the integrated charge $\Delta Q$ appears at $\tau_1 = \tau_2$ for all spin resonance lines, while no further features are resolved up to 900 ns. The features at $\tau_2 \lesssim 60$ ns are so-called Ramsey fringes, which are observable for evolution periods $\tau_2$ below the dephasing time of the system. A $\tau_{\pi/2}$ of 31 ns was used, and the experiment was performed at 6.5 K.
FIG. 3: Electrically-detected Carr-Purcell echoes observed on the isolated high-field P resonance at $B_0 = 351.2$ mT for different evolution times $\tau_1$. The solid line represents a mono-exponential decay with a timeconstant of $1.7 \pm 0.2 \, \mu s$. 

$\tau_{\text{echo}} = 1.7 \pm 0.2 \, \mu s$