Electrical Detection of Spin Coherence in Silicon

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Experimental evidence is presented showing that photocurrents in silicon can be used as highly sensitive readout probes for coherent spin states of localized electrons, the prime candidates for quantum bits in various semiconductor based quantum computer concepts. Conduction electrons are subjected to fast Rabi oscillation induced by means of pulsed electron spin resonance. The collective spin motion of the charge carrier ensemble is reflected by a spin-dependent recombination rate and therefore by the sample conductivity. Because of inhomogeneities, the Rabi oscillation dephases rapidly. However, a microwave induced rephasing is possible causing an echo effect whose intensity contains information about the charge carrier spin state and the coherence decay.

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In recent years, electron spins have played an increasing role in new concepts of semiconductor based quantum computers (QC) [1,2] that appear to have prospects for an experimental implementation [3]. These concepts utilize electron spins either as quantum bits (qubits) or as negotiators between nuclear spin qubits. Common to all concepts is that the qubit readout requires the measurement of coherent states of single or very small numbers of electrons. The classic experiment for the electron spin measurement is electron spin resonance (ESR), the spectroscopy of spin eigenstates separated by magnetic field induced Zeeman splitting [4]. For the observation of coherent spin motion, modern pulse ESR has to be used, which utilizes the Rabi nutation induced by strong resonant microwave fields (B_1) [5] for the preparation of arbitrary noneigenstates. The subsequent, coherent propagation of a spin ensemble is then detected indirectly through the emitted radiation induced by the weak polarization (Zeeman separation in μeV range). Consequently, pulse ESR is very insensitive for the readout of small numbers of spins and, therefore, not feasible for QC readout concepts. Various theoretical approaches have been developed for the solution of this problem [6–8], which are different for the different QC concepts, but commonly take advantage of spin-selection rules of electronic transitions. This principle has the advantage that transitions, which are energetically much larger than spin transitions, allow a much higher detection sensitivity while the information about the spin state to be measured is encoded in their spin dependency. In the past, such indirect spin-measurement techniques have been demonstrated on optical transitions. For instance, a combination of cw ESR with optical single-molecule detection allows the observation of the magnetic resonance of a single spin [9,10]. For the readout of solid state QCs in low band gap materials like silicon, these optically detected magnetic resonance techniques are not feasible since either the spin-dependent transitions are nonradiative or radiative at wavelengths too long to be detected at very low intensities and high time resolution. Hence, existing readout concepts are based on the measurement of spin-controlled charge currents such as in single spin transistors as proposed by Kane [7] and Vrijen *et al.* [8] for a silicon based nuclear spin QC or through charge currents in quantum dots as proposed for a QC concept by Recher *et al.* [6]. These concepts require that coherent spin motion is reflected by the charge currents. Information contained in spin states can be extracted only when the readout transition takes place within the coherence times.

In the following, experimental evidence for an imprint of coherent spin motion on a semiconductor current is presented. It is shown that dephasing and rephasing of Rabi oscillation in charge carrier ensembles can be reflected by a sample conductivity. The idea behind the experiment is to use pulsed ESR for the induction of Rabi oscillation of paramagnetic recombination centers which are localized, recombination active singly occupied electronic band gap states. The oscillation of the spin state will be reflected by spin-dependent recombination transitions, and, therefore, it can be detected by means of transient photoconductivity measurements.

The material for the experiments has to meet several requirements: First, a semiconductor is needed which provides microscopic point defects that introduce localized band gap states similar to those proposed for the mesoscopic readout devices of the quantum computer concepts mentioned above [1,2,6,7]. Second, spin-orbit coupling within the material has to be weak such that spin conservation imposes spin-selection rules on electronic transitions. In addition, control of the mutual exchange between these defects is necessary in order to prevent fast decoherence due to spin-spin relaxation. Because of this, silicon in its microcrystalline morphology is chosen as a model semiconductor. Crystalline silicon is proposed as host material for several quantum computer readout concepts [1,7,11]. Spin-orbit interaction is weak in all existing silicon morphologies, and many spin-dependent transitions are known [12–16] as well. In the microcrystalline morphology, localized paramagnetic deep band gap states due to silicon dangling bonds

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(db) exist, whose density, and hence their mutual interaction, is controllable by means of growth parameters [17].

Spin-dependent recombination of conduction electrons ($e-$) and holes ($h+$) at db centers in silicon works as illustrated in Fig. 1 [12,18–21]: When $e-$ localize close to the paramagnetic db states 1(a), strongly coupled intermediate electron pair states 1(b) are generated randomly in one of four possible spin eigenstates, the two symmetric triplet states $|T+\rangle$ and $|T-\rangle$ as well as in the triplet state $|T_0\rangle$ and singlet state $|S\rangle$. In the presence of a magnetic field, the latter two states are not purely symmetric and antisymmetric, respectively, but tilted slightly toward each other. This is due to the finite ratio between the spin exchange interaction and the nonvanishing difference of the Zeeman splitting within the pair. Since adjustment transitions 1(c) into the energetically lower charged db^- state are more favorable for singlet states, the adjustment time, and therefore the recombination time, is long for the triplet states $|T+\rangle$ and $|T-\rangle$ and shorter for the two mixed eigenstates. Thus, while $e-$ localized in $|T+\rangle$ and $|T-\rangle$ pairs are more likely to dissociate back to the conduction band without recombination 1(d), $e-$ in $|T_0\rangle$ and $|S\rangle$ pairs are likely to adjust into the db^- quickly, and, therefore, they will recombine fast with $h+$ from the valence band 1(e).

The experiment is started from a steady state photocurrent. Because of the constant pair generation and recombination under this condition, pure triplet states $|T+\rangle$ and $|T-\rangle$ are pumped to very high densities compared with the short lived eigenstates with singlet content. When a coherent ESR radiation that is polarized along an axis perpendicular to the magnetic field is switched on, Rabi oscillation between $|T+\rangle$ and $|T-\rangle$ states and the $|T_0\rangle$ state begins. Because of the higher recombination probability of the $|T_0\rangle$ state, the recombination rate increases each time a pair passes the $|T_0\rangle$ state. Hence, the Rabi oscillation leads to an oscillating recombination rate. After the pulse, a slow photocurrent relaxation takes place that is dependent on the pair ensemble state in the moment when the pulse ends. This allows one to obtain information about the dynamics of the spin-pair ensemble during the pulse by means of a current measurement at a given time $t_{\mu s}$ after the end of the pulse as a function of the pulse length τ. The theoretical details of this measurement approach are outlined elsewhere [20,21].

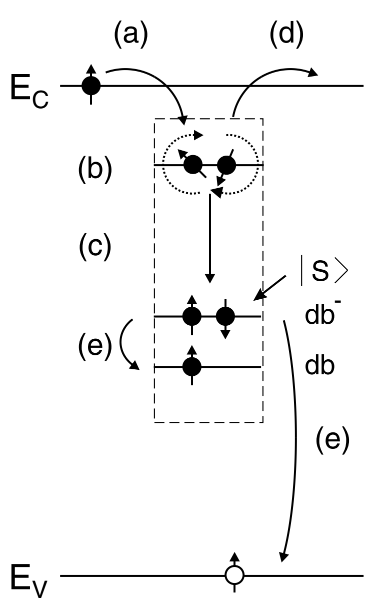

FIG. 1. Spin-dependent recombination and its coherent manipulation (dashed arrows) at a db center illustrated for an energy band diagram. The charged db^- ground state can exist only as a singlet state $|S\rangle$. Since spin conservation is present, transitions from other spin configurations have highly spin-dependent transition probabilities. For details see text.

Experimentally, the coherent spin manipulation is introduced by a Bruker E580 pulsed ESR spectrometer. The sample was exposed to a constant homogeneous magnetic field at about $B_0 = 347$ mT such that the db with Landé factor of $g \approx 2.005$ was brought into ESR. In order to verify that the observed signals are due to the ESR induced spin motion of charge carriers and not due to microwave artifacts or magnetoresistance effects, a magnetic field sweep was recorded which showed a transient response at the db resonance only [22]. The photocurrent was measured at $t_{\mu s} = 20$ μs. The microwave frequency was $\nu = 9.751\,972$ GHz (X band). The pulse length was changed in 2 ns steps. For the coherent amplification of the microwave radiation, a 1 kW traveling wave tube amplifier was used, yielding microwave field strengths in the lower mT range. The sample is a 2.7 μm thick film of hydrogenated microcrystalline silicon, deposited on 1737 Corning glass by electron-cyclotron resonance chemical vapor deposition as outlined elsewhere [23]. It was contacted with 48 interdigited aluminum grids of 300 nm thickness, 30 μm width, and 50 μm distance. In order to pump the $|T+\rangle$ and $|T-\rangle$ pair states, a steady state photocurrent ($I_{ph} = 100$ μA) was established under illumination of a 0.021(4) cm^2 sample area by Ar^+-ion laser light with $\lambda = 514$ nm wavelength and $P = 600$ mW intensity. The sample temperature was $T = 10$ K.

The experimental results are displayed in Figs. 2 and 3. Figure 2 shows a fast decrease of the photocurrent (increase of recombination) right after the beginning of the ESR pulse at $\tau = 0$. The photocurrent saturates after about $\tau = 30$ ns. In the data set given, it is hard to recognize an oscillatory behavior. Thus, it is difficult to determine whether the displayed data are actually due to fast dephasing Rabi oscillation, as expected, or due to an incoherent relaxation of the recombination rate to an on-resonant steady state value. The Rabi frequency $\Omega = \sqrt{(\omega_L - \omega)^2 + \gamma B_1^2}$ can be distributed broadly, due to the inhomogeneous distribution of the Larmor frequency ω_L (around the microwave frequency $\omega = 2\pi\nu$) as well as to a distribution of the microwave field strength

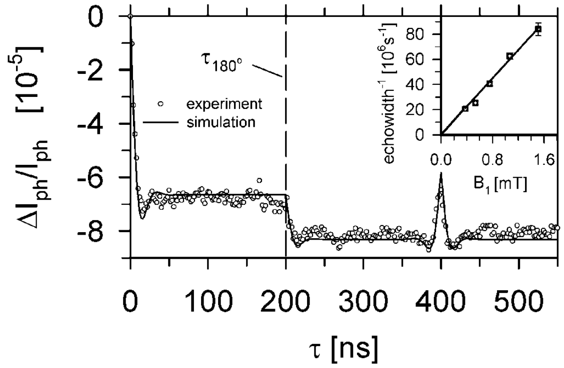

FIG. 2. The relative photocurrent change $\frac{\Delta I_{ph}}{I_{ph}}$ as a function of the pulse length τ measured at the db resonance. The microwave phase change at $\tau_{180°} = 200$ ns leads to a steplike dephasing process. At $\tau = 2\tau_{180°}$, an echo is observed. The solid line is the simulation of the experiment assuming strong coupling within the spin pairs. The inset illustrates the proportionality of the inverse echo width (inverse of full width at half maximum) as a function to the microwave field strength B_1. The solid line is a linear fit through the origin.

B_1. The latter is caused by mode distortion in the microwave cavity due to the electrical wires and contacts. In order to prove that the transient observed originates from coherent spin propagation, an echo experiment has to be carried out. The idea behind such a "recombination echo" experiment is to rephase Rabi oscillation by means of a 180° phase change of the microwave field B_1. As shown in Fig. 2, when a sudden (on a ps second time range) 180° phase change is introduced at a time $\tau_{180} = 200$ ns, a photocurrent peak can be observed at $\tau = 2\tau_{180} = 400$ ns, but note that the dephasing cannot be fully recovered. Hence, even though a direct observation of the Rabi oscillation is not possible due to the distribution of the Rabi frequencies, it can be verified by means of a collective temporary rephasing. The steplike photocurrent decrease right after the microwave phase change is discussed below. To confirm the interpretation of the photocurrent phenomenon shown in Fig. 2 as a collective effect of coherent Rabi oscillation, its dependence on the microwave field strength B_1 was measured. Since $\Omega \propto B_1$ when $B_1 \gg \omega_L - \omega$, an antiproportionality of the echo width to the B_1 field should be present. As depicted in the inset of Fig. 2, the inverse echo width exhibits the predicted linear dependence on B_1, and thus, the echo effect can only be due to Rabi oscillation.

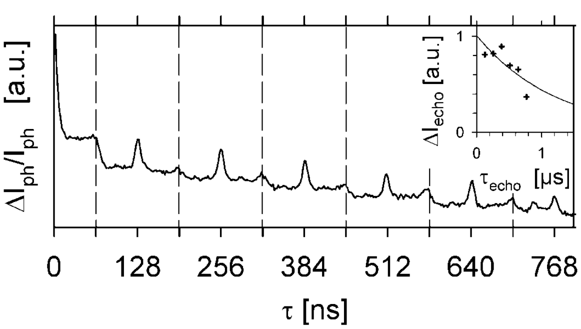

FIG. 3. The relative photocurrent change $\frac{\Delta I_{ph}}{I_{ph}}$ measured as a function of the pulse length τ at the db resonance when repeated phase changes are introduced as indicated by the vertical dashed lines. Note that the dephasing after the first phase change at $\tau = 64$ ns does not repeat when the other phase changes are introduced. The inset shows the echo peak intensity as a function of the time τ_{echo} when the echo occurred. The solid line is a single exponential function.

The electronic detection of coherent spin motion allows coherence time measurements. Since charge carrier pairs recombine during the microwave pulse, the magnitude of the echo should depend on the time τ_{echo} when it occurs. Hence, as long as the decoherence resulting in the decay of the recombination echo is not dominated by the longitudinal spin-lattice relaxation time T_1 or the transverse spin-spin relaxation time T_2, it reveals the recombination time of the db channel. In order to determine the echo decay with increasing τ_{echo}, an echo-echo experiment was carried out, whose result is depicted in Fig. 3. After the initial echo that follows the first phase change, additional phase changes are introduced leading to echoes of previous echoes. Note that there are only two dephasing signatures, one right after the pulse begins and one after the first phase change. Thereafter, the spin pairs that cannot be rephased have no contribution to the observed signals anymore and only the rephasable part of the ensemble determines the transient. Hence, echo echoes are always as strong as previous echoes except for a small decrease due to pair losses. One can see this slow decline of the echo intensity with increasing pulse length in the inset of Fig. 3. An exponential fit of this decline reveals a time constant of $\tau_f = 1.3(5)$ μs.

The experimental data presented above can be understood by comparison with the simulation of the photocurrent as displayed by the solid line in Fig. 2. It is based on a model for coherent, ESR induced Rabi oscillation of localized electron hole pairs with Gaussian distributed Rabi frequencies. Exchange coupling between the pair partners is assumed to be strong due to small g-factor separation; influences due to incoherence are considered to be negligibly small. Note that it is the strong coupling that causes the steplike dephasing after the phase change. Details about the model and its validity for the simulation of recombination rates are discussed elsewhere [20]. The simulation was fit to the experimental data with two fit parameters: (i) A vertical current scaling factor that is proportional to the arbitrary sample conductance, and (ii) the width of the echo. This means that the ratios between the signal magnitude before or after the second dephasing and the echo height are not fit but solely determined by theory. Thus, the excellent agreement of the simulation and the measured data is another strong indication for the correctness of the interpretation of the observed phenomena as current imprints of spin-Rabi oscillation.

The result of the echo-echo decay measurement has two remarkable implications: First, the process responsible for the decay is not spin relaxation since T_1 and T_2 times, measured by conventional pulse ESR [21,24], are more than an order of magnitude slower in the given sample. Thus, the decay can be attributed to the loss of charge carrier pairs due to recombination transitions. Second, the τ_f decay is the fastest source of incoherence since any faster source would lead to a faster echo decay. This shows that spin coherence times of localized charge carrier pairs in silicon can be many orders of magnitude longer than the electronic coherence time of conduction electrons in the same material [25].

The experiments presented are in principle applicable to any material where charge carrier transport or recombination involves spin-dependent transitions through localized states at point defects or artificial structures such as quantum dots or wells. This shows that the measurement of spin coherence by means of charge currents as proposed theoretically by Engel and Loss [6,26] is a feasible approach. The experiments shown here are also important for semiconductor characterization in general. Quantitative information about distinct recombination and other charge carrier transitions in semiconductors is accessible on a microscopic level. For the detection setup used [27], a detection limit for relative photocurrent changes of 10^{-7} was possible. At the maximum time resolution this allows the detection of only ≈ 100 recombining electron hole pairs which is about 9 orders of magnitude more sensitive than conventional ESR spectroscopy. Note that this is achieved in the presence of the strong steady state offset photocurrent. In the absence of this current, whose only purpose is to pump the high triplet density in order to initialize the spin ensemble, even higher sensitivities can be achieved. Hence, semiconductor charge carrier currents can be used as probes for spin states of small electron spin ensembles, and, therefore, they are potential observables for readout mechanisms of semiconductor based spin quantum computers.

In conclusion, we have given experimental evidence for the existence of coherent spin motion effects on charge carrier currents in a semiconductor. The temporal and microwave intensity dependence of these phenomena as well as the possibility of echo echoes confirm that they are caused by the dephasing and rephasing of Rabi oscillation of localized charge carriers in db states of silicon. The importance of these findings with regard to application for highly sensitive readout devices of solid state quantum computers and material characterization have been outlined.

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