Spin Manipulation of Free Two-Dimensional Electrons in Si/SiGe Quantum Wells

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Understanding the mechanisms controlling the spin coherence of electrons in semiconductors is essential for designing structures for quantum computing applications. Using a pulsed electron paramagnetic resonance spectrometer, we measure spin echoes and deduce a spin coherence time (T_2) of up to 3 μs for an ensemble of free two-dimensional electrons confined in a Si/SiGe quantum well. The decoherence can be understood in terms of momentum scattering causing fluctuating effective Rashba fields. Further confining the electrons into a nondegenerate (other than spin) ground state of a quantum dot can be expected to eliminate this decoherence mechanism.

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Currently there is considerable interest in the possibility of using the spin of an electron in a semiconductor as a qubit for quantum information processing [1]. Silicon appears to be a particularly promising host because the spin-orbit interaction is small, the dominant naturally occurring isotope has no nuclear spin, and the technology of Si integrated circuits is so well developed. It has been known for 40 years that electrons bound to donors in Si have spin-population lifetimes of hours, and coherence times approaching a millisecond [2]. Recent results have demonstrated coherence times of at least 60 ms [3]. However, these long relaxation times are observed with isolated bound electrons in a homogeneous, bulk semiconductor. Some of the structures envisioned for semiconductor spin-based quantum information processing make use of the well developed technology of heterostructures and involve drawing the electrons to a heterointerface where they are controlled by electrostatic gates [1,4]. However, there have been no reports of direct spin coherence measurements for electrons at a heterointerface.

Here we report the first spin-echo measurements of the spin coherence time (T_2) of two-dimensional (2D) electrons in a semiconductor heterostructure. The electrons form a high mobility 2D electron system in a one-side modulation-doped Si quantum well embedded in SiGe, similar to structures studied earlier by conventional continuous wave (cw) EPR [5–7] and electrically detected magnetic resonance (EDMR) [8]. Microwave pulses in a pulsed-EPR spectrometer produce the necessary spin rotations (which can be viewed as elementary single-qubit operations) with two- and three-pulse sequences being used to measure the transverse relaxation (decoherence or phase memory) time, T_2, and the longitudinal (spin-population) relaxation time, T_1. The longest measured coherence times are $T_2 \sim 3$ μs, with longitudinal times of $T_1 \sim 2.3$ μs. These times are $>10^4$ times the spin precession period. The fact that $T_2 > T_1$ is unusual and indicates that the relaxation processes are anisotropic [9,10]. The relaxation-time measurements are consistent with the suggestion, based on extensive cw EPR experiments [7], that the longitudinal relaxation time and coherence time are controlled by effective in-plane fluctuating magnetic fields, or Rashba fields [11], arising from the spin-orbit interaction and the broken inversion symmetry at the heterostructure interface. We deduce a Rashba field of 10–20 G, orders of magnitude smaller than in some III-V systems [12], and observable only because of the long inherent spin relaxation times in Si. These results suggest several ways in which the spin decoherence may be reduced: lateral confinement into quantum dots or wires, more symmetric quantum wells, and using larger magnetic fields and microwave frequencies.

The samples used for these experiments are one-side modulation-doped quantum wells grown by molecular beam epitaxy , with a 0.5 μm strain-relaxed $Si_{0.75}Ge_{0.25}$ buffer atop a 2–2.5 μm compositionally graded $Si_{1-x}Ge_x$ layer, followed by the Si quantum well, doped $Si_{0.75}Ge_{0.25}$ layers and a Si cap. The details of the growth and electrical measurements have been described elsewhere [8]. Most of the experiments reported here were performed on a structure (sample 1) with a 20 nm quantum well, an electron density, $n \sim 3 \times 10^{11}/\mathrm{cm}^2$, and a mobility, $\mu \sim 9 \times 10^4$ $\mathrm{cm}^2/\mathrm{Vs}$, after illumination [6,13]. A second structure (sample 2) with a 15 nm quantum well and improved transport properties ($\mu \sim 190\,000$ $\mathrm{cm}^2/\mathrm{V\,s}$, $n \sim 1.7 \times 10^{11}/\mathrm{cm}^2$ [14]) has also been studied. The modulation doping density was kept low in sample 1, and little or no EPR signal is observed initially. However, electrons can be introduced into the quantum well by illumination and persistent photoconductivity [5]. In our experiments we have first cooled the samples in the dark and then illuminated them for a sufficient time to reach saturation, as measured by the cw EPR signal level. The illumination was provided by a room temperature GaAs light emitting diode with its peak emission at about 900 nm, and brought to the sample through a window in the cryostat and cavity. Sample 2 does not show significant persistent photoconductivity, and it was always cooled and maintained in the dark.

Measurements were performed with a Bruker Elexsys580 X-band EPR spectrometer using a dielectrically loaded cylindrical resonator (EN-4118MD4). The samples were held in a fused silica tube, immobilized with frozen ethanol, and the entire cavity and sample maintained at low temperature (4–5 K) with a helium-flow cryostat (Oxford CF935). The temperature was controlled to better than 0.05 K with a calibrated Cernox temperature sensor, though no significant temperature dependence was found over the range from 3.5 to 8 K.

A two-pulse Hahn echo experiment ($\pi/2 - \tau - \pi - \tau -$ echo) was used to measure T_2 (a detailed explanation, including various pulse sequences, can be found in Ref. [15]). In our two-pulse experiments the microwave frequency was offset by 7–8 MHz from the resonance line to minimize interference from background EPR signals from the cavity. This offset results in an oscillating echo shape. Both the in-phase (real) and quadrature (imaginary) components of the decaying portion of the echo signal were detected and accumulated with a transient recorder. The first ($\pi/2$) and second (π) pulse durations were 16 and 32 ns, respectively, which corresponds to an excitation bandwidth of 16 MHz (larger than the frequency offset). Representative signals for sample 1 are shown in Fig. 1(a) for the case where the applied magnetic field is perpendicular to the 2D electron system ($\theta = 0$). A 16-step phase-cycle scheme was applied to eliminate unwanted signals, in particular, the free-induction decay (FID) arising from each of the individual microwave pulses. The raw data are Fourier transformed, producing an EPR line which is shifted from the carrier frequency by 7–8 MHz. In Fig. 1(b) we show these Fourier transformed (FT-EPR) spectra for two interpulse delays. The integrated intensity of the FT-EPR lines for different τ and at $\theta = 0$ are plotted in Fig. 2(a) (plotted against the total time, 2τ) for an as-illuminated sample (solid squares) and after a 30 K "anneal" (solid circles). Annealing to 30 K for a few minutes after illumination resulted in a decrease in the cw EPR linewidth (from about 150 to 60 mG), while leaving the integrated EPR intensity essentially unchanged. We find that T_2 is correspondingly increased. The curves shown in Fig. 2(a) are exponential fits to the data, giving $T_2 = 0.96$ and 2.98 μs before and after annealing, respectively. The fitted curves do not pass through all the data points, especially at longer delays, indicating that there is a distribution of phase memory times.

Sample 2 with a higher mobility and a lower electron density gave similar results, although no illumination and annealing steps were necessary. The EPR line in this structure appears to be homogeneously broadened; no distinct echo signal was observed, and therefore the coherence time is given by the free-induction decay time ($T_2{}^*$), $T_2 = T_2{}^* = 1.4$ μs.

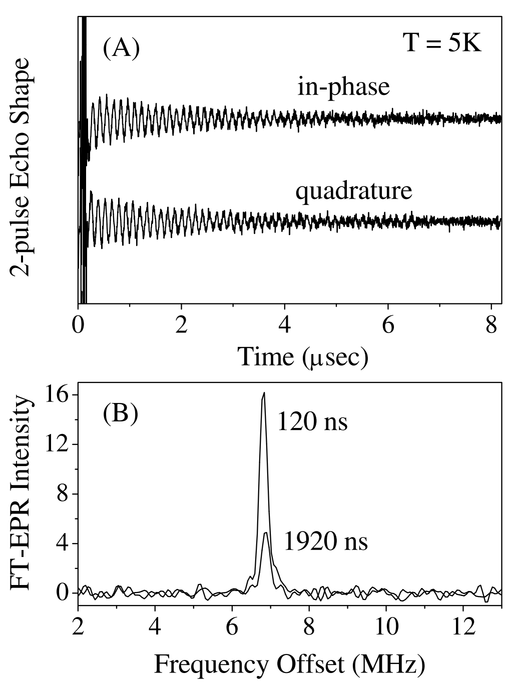

FIG. 1. (a) Oscillating shape of the in-phase and quadrature echo signals in a two-pulse spin-echo experiment after the 30 K anneal at $\theta = 0°$. Only the decaying portion of the echo shape is shown with the echo center at 0.1 μs (the echo rise and early decay are obscured by the cavity ring down and detector blanking). (b) Fourier-transform EPR lines for two interpulse delays, τ. The integrated intensity of these FT-EPR lines versus delay is used to measure T_2.

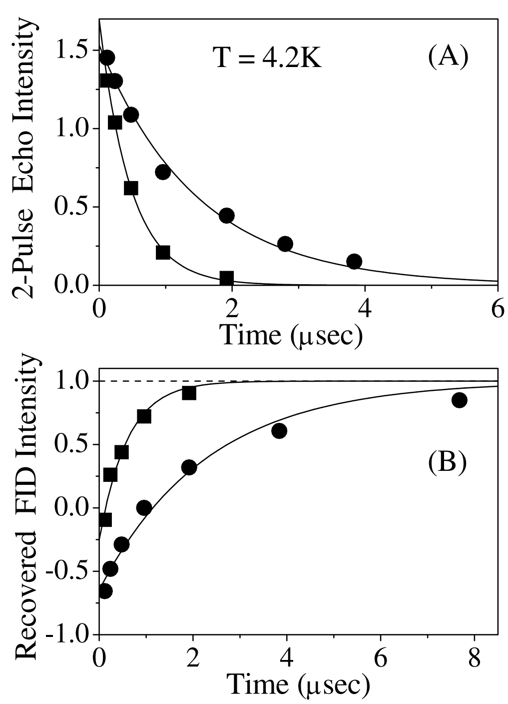

FIG. 2. (a) Two-pulse spin-echo intensity versus total delay time (2τ), and (b) FID intensity in the inversion-recovery experiment as a function of interpulse delay T, for the as-illuminated (■) and after 30 K anneal (●) with $\theta = 0°$. The exponential fits give T_2 in (a) and T_1 in (b).

We also performed a two-pulse inversion-recovery experiment ($\pi - T - \pi/2 -$ FID) to measure the longitudinal relaxation time, T_1. Similar to the T_2 experiment, the microwave frequency was offset by 7–8 MHz from the resonance line and the entire oscillating FID was accumulated by the transient recorder. This FID signal was Fourier transformed and the integrated intensity of the recovered EPR line was analyzed as a function of delay, T, to extract the longitudinal relaxation time T_1. An eight-step phase-cycle scheme was used to eliminate unwanted signals: the FID from the first microwave pulse and the echo signal originating from the two pulses. For θ near 90° the EPR lines are broad and weak so that the short FID is lost in the cavity ring down. To avoid this problem a third (π) microwave pulse at the end of the sequence was used to produce an oscillating echo, which is more easily detected because it is well separated in time from the applied microwave pulses. The resulting three-pulse sequence is $\pi - T - \pi/2 - \tau - \pi - \tau -$ echo, with τ held constant at 100 ns. The results (sample 1) for T_1 are shown in Fig. 2(b), with the solid squares from the as-illuminated sample and the solid circles after the anneal. The curves in Fig. 2(b) are exponential fits to the data which show $T_1 = 0.6\ \mu$s in the preanneal experiments and $T_1 = 2.0\ \mu$s postanneal ($\theta = 0$). As with T_2, from the quality of the fits we see that there is a distribution of spin relaxation times. Sample 2 had a somewhat shorter $T_1(0.95\ \mu\text{s})$, and it also showed a distribution of relaxation times.

It is particularly striking that $T_2 > T_1$ in these structures. This is an unusual situation and requires the relaxation processes to be anisotropic. From an abstract perspective, any process leading to relaxation or decoherence of a spin can be viewed as a fluctuating magnetic field acting on the spin. In the Redfield limit ($\gamma \delta B \tau_c \ll 1$, where γ is the electron gyromagnetic ratio, and τ_c is the correlation time of the fluctuations) and assuming that the fluctuating fields, δB, along different spatial axes are uncorrelated, the relaxation times are given by [10]

$$\frac{1}{T_1} = \gamma^2(\overline{\delta B_x^2} + \overline{\delta B_y^2})\frac{\tau_c}{1 + \omega_0^2\tau_c^2},$$

$$\frac{1}{T_2} = \gamma^2\overline{\delta B_z^2}\tau_c + \frac{1}{2T_1},$$

where the external magnetic field, B_0, is assumed to be applied along the z direction and ω_0 is the Larmor frequency of the spin in this field. If the fluctuating fields are isotropic, T_1 is always greater than or equal to T_2. On the other hand, $T_2 = 2T_1$ if $\overline{\delta B_z^2} = 0$ and $\overline{\delta B_x^2}, \overline{\delta B_y^2} \neq 0$ [10]. This situation has been observed in other anisotropic systems [9].

Based upon cw EPR measurements it has been suggested [7] that the spin relaxation of the 2D electrons in these Si quantum well structures is controlled by fluctuating effective magnetic fields (Rashba fields [11]) arising from the breaking of inversion symmetry by the Si/SiGe interface and the electric field in the quantum well. The Rashba fields lie entirely in the plane of the 2D electron system, resulting in $\overline{\delta B_x^2}, \overline{\delta B_y^2} > \overline{\delta B_z^2}$, which can lead to T_2 being longer than T_1. The correlation time of the field fluctuations, τ_c, should correspond approximately to the electron momentum relaxation time which is about 10 ps for $\mu \sim 90\,000\ \text{cm}^2/\text{V s}$ (sample 1). Using this value for τ_c we estimate fluctuating in-plane fields, δB_x, $\delta B_y =$ 10.5 G, and the out-of-plane field fluctuations to be $\delta B_z =$ 5 G to fit $T_1 = 2.3\ \mu$s and $T_2 = 3\ \mu$s measured after the 30 K anneal. The relaxation times are shorter before the anneal ($T_1 = 0.6\ \mu$s and $T_2 = 0.96\ \mu$s) and for the same τ_c correspond to fields of 19 G in plane and 8 G in the z direction. These field values are only estimates, since it was not possible to measure the mobility of the 2D electrons at the same time and under the same conditions as the pulsed EPR.

The longer scattering time in sample 2 (about 22 ps for $\mu \sim 190\,000\ \text{cm}^2/\text{V s}$) implies less averaging of the Rashba fields (similar to motional narrowing) and a shorter $T_2(\omega_0\tau \sim 1.4)$. To fit the measured relaxation times, we find $\delta B_x, \delta B_y = 14.5$ G and the out-of-plane field fluctuations to be $\delta B_z = 5$ G. Samples 1 and 2 have similar in-plane and out-of-plane contributions to their spin relaxation.

We can directly test for the anisotropy expected from the Rashba effect by measuring the spin relaxation times as a function of the angle, θ, of the 2D electron system with respect to the applied magnetic field. If the sample is tilted, part of the in-plane Rashba field will now contribute to δB_z (with the z direction defined by B_0). Thus we expect T_2 to decrease while T_1 should increase for $\theta \neq 0$. In Fig. 3 we show T_1 (solid squares) and T_2 (solid circles) for different angles of sample 1 with respect to the external magnetic field. As expected, T_2 drops while T_1 rises. A similar angular dependence is found in sample 2, and the resonance becomes inhomogeneously broadened ($T_2 > T_2{}^*$)

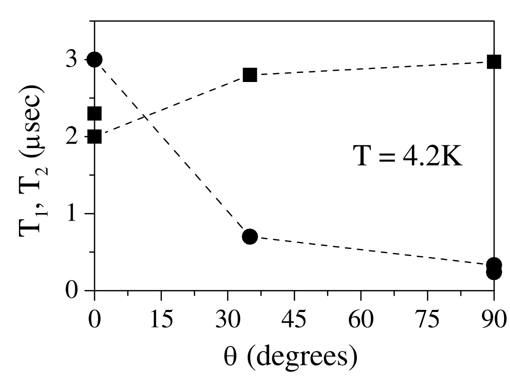

FIG. 3. Electron spin relaxation times, T_1 (■) and T_2 (●), of the 2D electron system (after the 30 K anneal) as a function of the angle (θ) between the external magnetic field, B_0, and the normal to the 2D electron system. The lines connecting experimental points are guides to the eye. The multiple values shown for T_1 at 0° and T_2 at 90° were obtained in different experimental runs, showing about 10% variations.

away from $\theta = 0$. Given the uncertainty in the correlation time, and how it might be affected by changing the orientation of the sample in the magnetic field, we have not attempted to fit the angular behavior of the relaxation times. However, the effect of rotating the sample is qualitatively consistent with what one would expect from the Rashba effect.

These measurements have important implications for devices utilizing electrons at heterointerfaces for quantum computing. While we have shown that a large number of spin operations can be performed within the spin coherence time, longer coherence will be advantageous for quantum computation. Even for architectures in which the electron remains bound in the bulk Si most of the time, the spin will be subject to an increased spin-orbit interaction (Rashba effect) during the time it is at an interface. There are several approaches to reducing the decoherence. Since the magnitude of the Rashba field is independent of the applied field, at sufficiently high microwave frequencies ($\omega_0 \tau_c \gg 1$) the effect of the Rashba fields will be suppressed and longer spin coherence times will ensue. Higher frequencies will also, in principle, allow shorter microwave pulses (faster qubit operations). A second approach is to reduce the Rashba field by increasing the symmetry of the potentials seen by the electrons. Both structures we have studied are one-side modulation doped, and thus have the maximum asymmetry. From a materials standpoint, symmetric structures with comparable mobilities are more difficult to fabricate, but have been grown [16].

A third approach is to laterally confine the electrons into quantum dots and quantum wires at the interface, rather than allowing them to move freely. If the electron travels ballistically along a quantum wire, the spin will be rotated by the Rashba field, but in a controlled and reversible manner. In the nondegenerate (other than spin) ground state of a quantum dot the electron will occupy a stationary state, and the Rashba fields will change its effective g tensor but will not affect the spin relaxation times. Excited states of the quantum will have different g tensors, and transitions between states will cause decoherence. Thus, the quantum dots must be held at a low enough temperature to freeze the electron into the ground state. Decoherence in the ground state can be caused by distortions of the wave function from time-varying fields, thereby mixing in excited states with a higher angular momentum. In most of the electron spin-based quantum computing proposals, such distortions are required to control the exchange coupling of two spins. However, it has been shown that the first-order effects can be eliminated by tailoring the time dependence of the distortion [17]. Uncontrolled distortions, from defect charge fluctuations, for example, must be minimized.

In summary, we have performed the first pulsed-EPR experiments (or equivalently, ensemble single-qubit operations) on free 2D electrons at a heterointerface. The spin-echo experiments give definitive measurements of electron spin coherence time in this system. We find relaxation times as long as 3 μs, about 30 000 times the microwave period. While these coherence times are relatively long, strong 3D confinement of the electron will be necessary if much longer coherence times (approaching that of donor electrons) are required. The large anisotropy in T_2 and the fact that $T_2 > T_1$ for the field perpendicular to the 2D electron layer imply that the processes giving rise to the spin relaxation can be viewed as effective fluctuating magnetic fields lying in the plane of the electrons. These results are consistent with the recent suggestion that the spin relaxation is being caused by Rashba fields arising from the spin-orbit interaction and the broken inversion symmetry in the quantum well [7]. We deduce Rashba fields of about 10–20 G in these Si/SiGe structures.

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