 Ultraslow Electron Spin Dynamics in GaAs Quantum Wells Probed by Optically Pumped NMR

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Optically pumped nuclear magnetic resonance (OPNMR) measurements were performed in two different electron-doped multiple quantum well samples near the fractional quantum Hall effect ground state \(\nu=\frac{1}{3}\). Below 0.5 kelvin, the spectra provide evidence that spin-reversed charged excitations of the \(\nu=\frac{1}{3}\) ground state are localized over the NMR time scale of about 40 microseconds. Furthermore, by varying NMR pulse parameters, the electron spin temperature (as measured by the Knight shift) could be driven above the lattice temperature, which shows that the value of the electron spin-lattice relaxation time \(\tau_{1L}\) is between 100 microseconds and 500 milliseconds at \(\nu=\frac{1}{3}\).

A two-dimensional electron system (2DES), cooled to extremely low temperatures in a strong magnetic field, exhibits many exotic phenomena, such as the fractional quantum Hall effect (FQHE) \([3]\). Transport and optical studies of the 2DES have shown that the low-energy physics in these extreme conditions is driven by the electron-electron Coulomb interaction \([4]\), but the challenge of precisely describing the low-lying many-body states that exist in a real 2DES remains formidable for both theory and experiment \([5,6]\). Such 2DESs have been probed by OPNMR \([6,7,8]\) in the FQHE regime \([9]\), which allows the direct radio-frequency (rf) detection of NMR signals from nuclei in electron-doped GaAs quantum wells. The \(^{71}\)Ga OPNMR spectra reveal the local, time-averaged value of the electron spin magnetization, \(\langle S_z (\vec{R}) \rangle\), thus leading to insights about the many-electron states relevant for the FQHE.

We report evidence of ultraslow electron spin dynamics near the most studied FQHE ground state, \(\nu=\frac{1}{3}\), with characteristic time scales exceeding \(\sim 40 \mu s\) below 0.5 K. Although the samples are characterized by “simple” NMR parameters (that is, isotropic hyperfine coupling in an oriented single crystal), the OPNMR spectra are complex because they can simultaneously exhibit inhomogeneous broadening due to the quantum confinement of electrons within a well and motional narrowing due to delocalization of electrons along the well \([10]\). At low temperatures \((T \approx 0.5\) K\), a change in the NMR linewidth is observed whenever spin-reversed electrons are present. We attribute this striking behavior to the localization of spin-reversed electrons over the NMR time scale. In addition, using the Knight shift as a thermometer, we measured the increase of the electron spin temperature above the lattice temperature when rf pulses were used to drive the system out of equilibrium. These non-equilibrium measurements imply that the electron spin-lattice relaxation time is \(100 \mu s < \tau_{1L} < 500\) ms for \(T < 0.5\) K at \(\nu=1/3\), which appears to exceed all electronic time scales previously measured in semiconductors by at least a factor of 1000.

Both of the multiple quantum well samples in this study were grown by molecular beam epitaxy on semi-insulating GaAs(001) substrates. Sample 40W contains 40 300 Å wide GaAs wells that were separated by 3600 Å wide Al\(_{0.1}\)Ga\(_{0.9}\)As barriers. Sample 10W contains 10 260 Å wide wells that were separated by 3120 Å wide barriers. Silicon delta-doping spikes located at the center of each barrier provided the electrons that were confined in each GaAs well at low temperatures, producing a 2DES with very high mobility \((\mu > 1.4 \times 10^6\) cm\(^2\)V\(^{-1}\)s\(^{-1}\)) \([11]\). The 2D electron densities in each well were \(n_{40W}=6.69 \times 10^{10}\) cm\(^{-2}\) and \(n_{10W}=7.75 \times 10^{10}\) cm\(^{-2}\).

The low-temperature \((0.29 K < T < 1.5 K)\), high-field \((B_{tot}=12\) T\) OPNMR measurements were performed with an Oxford Instruments sorption-pumped \(^{3}\)He cryostat that was mounted in a Teslatron\(^{12}\) superconducting magnet. The samples, about 4 mm by 6 mm by 0.5 mm, were in direct contact with helium and were mounted on the platform of a rotator assembly in the NMR probe. By tilting this platform, we could vary the angle \(\theta (-60 ^\circ < \theta < 60 ^\circ, \pm 0.1 ^\circ)\) between the sample’s growth axis \(\vec{z}\) (perpendicular to the plane of the wells) and the applied field \(\vec{B}_{tot}\) (fixed along \(\vec{z}\)), thereby changing the filling factor \(\nu=\left(nhc/(eB_{tot} \cos \theta)\right)\) in situ (here \(B_{tot}=B_{tot} \cos \theta)\), where \(h\) is Planck’s constant, \(c\) is the speed of light, and \(e\) is the electron charge. For optical pumping, light from a Coherent 890 Ti:Sapphire laser was delivered into the cryostat through an optical fiber \([12]\), which was terminated by a collimating lens and a polarizing assembly 22 cm above the sample. The light spot on the sample (5 mm diameter, 812 nm wavelength, left-circularly polarized, \(\leq 10\) mW cm\(^{-2}\) was gated by a spectrometer-controlled room-temperature shutter.

For the OPNMR measurements, we used the timing sequence SAT–\(\tau_L\)–DET \([6,7,8,9]\): SAT denotes an rf pulse train that destroys (saturates) the \(^{71}\)Ga nuclear po-
The dashed line fit is obtained by broadening the polarization throughout the sample, \( \tau_L \) is light time, \( \tau_D \) is dark time, and DET denotes the detection period. During \( \tau_L \) (30 to 90 s), optical pumping of interband transitions generated electrons and holes in the GaAs wells with nonequilibrium spin polarizations, which then polarized the nuclei in the wells through the hyperfine interaction. The shutter was then closed to allow the electrons to equilibrate with \( ^3\text{He} \) bath during \( \tau_D \) (typically 40 s). The enhanced nuclear polarization persisted until DET, whereupon a single rf tipping pulse was applied to produce a free induction decay signal, which we then acquired with a home-built NMR spectrometer that was based on a Tecmag Aries system. A calibrated RuO\(_2\) thermometer, in good thermal contact with the sample, recorded the temperature during signal acquisition.

A \(^{71}\text{Ga}\) OPNMR emission spectrum at \( \nu = \frac{1}{3} \) (Fig. A, solid line) exhibits a “w” peak that arises from nuclei in the GaAs quantum well and a “b” peak that is due to nuclei in the Al\(_{0.1}\)Ga\(_{0.9}\)As barriers. The Fermi contact hyperfine coupling between the spins of the 2DES and nuclei in the well shifts the w peak below the b peak by \( K_S \), which we define to be the Knight shift. The asymmetry of the well line shape has two origins: (i) the quantum confinement within the well causes the electron density to vary across its width \( w \) as \( \rho(z') \approx \cos^2(\pi z'/w) \) for \( |z'| \leq w/2 \) [4,12] and (ii) the optical pumping preferentially polarizes nuclei in the center of the well. Taking these two effects into account, the intrinsic line shape (Fig. 1A, hatched region) may be written as the sum of \( I_w^{\text{int}}(K_{\text{int}}, f) = [f/(K_{\text{int}} - f)]^{1/2} \) and \( a_0\delta(0) \) for the unbroadened barrier signal. Using a 3.5-kHz full width at half maximum (FWHM) Gaussian \( g(f) \) for the nuclear dipolar broadening [10], we arrive at a two-parameter fit (Figs. 1 and 2, dashed lines):

\[
I(f) = I_b + I_w = a_b g(f) + \int_0^{16} df' g(f - f') I_w^{\text{int}}(K_{\text{int}}, f')
\]

The first parameter, \( a_b \), is the amplitude of the barrier signal, which grows during \( \tau_L \) as the optically pumped nuclear magnetization diffuses out of the quantum well. The second parameter extracted from the fit is the hyperfine shift for nuclei in the center of the well, \( K_{\text{int}}(\nu, T) = \mathcal{P}(\nu, T) \frac{\nu}{S_z} (4.5 \pm 0.2) \times 10^{-16} \text{kHz cm}^3 \) [1]. Thus, fits to OPNMR spectra at various \( \nu \) and \( T \) provide a direct measure of the electron spin polarization \( \mathcal{P}(\nu, T) \equiv \langle S_z(\nu, T) \rangle / \langle S_z \rangle \), which is the charge-spin-texture excitations of the \( \nu = 1 \) ground state [4]. Recently, \( \mathcal{P}(\nu) \) was found to drop on either side of the \( \nu = \frac{1}{3} \) ground state, which shows that the charged excitations of this FQHE ground state are partially spin reversed, even in a 12-T field [1].

In all of these earlier measurements, the OPNMR spectra were well described by the dashed-line fits generated by our model. The central assumption of this model is that the electron spins are delocalized along the well, such that \( \langle S_z(\nu, T) \rangle \) appears spatially homogeneous when averaged over the NMR time scale (\( \sim 40 \mu s \)) [13]. In this limit, the delocalization of the low density 2DES (there are \( \sim 10^6 \) nuclei per electron in the well) produces a motional narrowing of \( I_w^{\text{int}} \).

However, low-temperature measurements at \( \nu = 0.267 \) showed a crossover to more complicated line shapes (Fig. 1B). Although the spectra were in reasonable agreement with our model above 1 K, the width of the w peak

FIG. 1. (A) The \(^{71}\text{Ga}\) OPNMR emission spectrum (solid line) of sample 10W at \( \nu = \frac{1}{3} \), taken at \( \theta = 36.8^\circ \) in \( B_{\text{ext}} = 12 \text{T} \). The frequency shift is relative to \( f_\nu = 155.93 \text{MHz} \). The dashed line fit is obtained by broadening the intrinsic line shape (hatched region). Empirically, \( K_{\text{int}} = K_S + 1.1 \times (1 - \exp(-K_S/2.0)) \) (in kilohertz). (B) Temperature dependence of the spectra at \( \nu = 0.267 \) (\( \theta = 0^\circ \)). The FWHM of the well resonance w peak is shown. Arb. units, arbitrary units.
increased dramatically as the temperature was lowered to $T=0.45\text{K}$ and then decreased upon further lowering to $T=0.31\text{K}$. This nonmonotonic temperature dependence is reminiscent of the behavior seen in NMR studies of systems in which spectra are sensitive to dynamical processes \[19\], variously referred to as motional narrowing, dynamical averaging, or chemical exchange \[12\]. In our experiment, the nuclei were rigidly fixed in the lattice of a single crystal, so the variation in the line shape shown in Fig. 1B was a signature of electron spin localization, which turned off the motional narrowing of the well resonance as the temperature was lowered.

The extra broadening of the well resonance disappeared as the sample was tilted from $\theta_{10\text{W}}=0^\circ$ ($\nu=0.267$) to $36.8^\circ$ ($\nu=1/3$) (Fig.2), despite a $10\%$ increase in the dipolar broadening (the $^{75}$As nearest-neighbors of the $^{71}$Ga nuclei are at the “magic angle” \[21\] when $\theta_{10\text{W}}=0^\circ$). Furthermore, there was a correspondence between the decrease in the linewidth and the increase in $K_S$ as $\nu\rightarrow1/3$ (Fig. 3). This anticorrelation strongly suggests that the behavior shown in Figs. 1 to 3 is due to electron spin dynamics.

For a quantitative understanding of these phenomena, we must consider the specific assumptions that lead to $I_w^{\text{int}}(K_{\text{Sint}}, f)$. Nuclei within the well couple to the spins of the 2DES through the isotropic Fermi contact interaction \[6\,7\,8\,9\,10\]; thus, a nucleus at site $\mathbf{R}'$ experiences a hyperfine magnetic field $\mathbf{B}_H(\mathbf{R}')=(16\pi\mu_B/3)\sum_j\mathbf{S}_j\delta(\mathbf{r}_j-\mathbf{R}')$, where $\mu_B$ is the Bohr magneton, $\mathbf{S}_j$ is the spin of electron $j$, the summation is over all of the conduction electrons within the well, and the delta function picks out those electrons that overlap with the nucleus at $\mathbf{R}'$. The average projection of $\mathbf{B}_H$ along the applied field $\tilde{B}_c$ may be written quite generally as $\langle \mathbf{B}_H(\mathbf{R}', \nu, T) \rangle = \langle \frac{8\pi\mu_B}{3} \rangle / (\langle |u(0)|^2 \rangle \langle |\chi(Z')|^2 \rangle \langle |\phi(X')|^2 \rangle) \mathcal{P}(\tilde{R}', \nu, T)$.

Here, the probability density of finding electrons at a $^{71}$Ga site has been factored into a term with the periodicity of the lattice $\langle |u(0)|^2 \rangle$ and terms that vary slowly within a unit cell $\langle |\chi(Z')|^2 \rangle \langle |\phi(X', Y')|^2 \rangle$ \[13\]. $\mathcal{P}(\mathbf{R}', \nu, T)$ is the local spin-polarization ($-1<\mathcal{P}<1$) of the electrons at $\mathbf{R}'$. If we assume that electrons are delocalized along the well, then the time-averaged values of $|\phi|^2$ and $\mathcal{P}$ are spatially homogeneous. In this limit, the local hyperfine frequency shift (taken to have the sign of $\mathcal{P}$) is a function of $\mathbf{z}'$ only, $\langle \mathbf{f}(\mathbf{z}') \rangle = \frac{-71\gamma \langle B_z'(\mathbf{z}', \nu, T) \rangle}{2\pi \cos^2(\pi z'/w) K_{\text{Sint}}}$, so the general expression for the well line shape is $I_w^{\text{int}}(f) = \int d^3\mathbf{r} \langle \mathbf{f}(\mathbf{r}) \rangle \rho_{\text{nuclear}} \delta(f-f(\mathbf{r}'))$. We further assume that wells are identical and that the optical pumping gives rise to a nuclear polarization that varies across each well as $\langle I_{\text{Sint}}(\mathbf{z}') \rangle \sim f(\mathbf{z}')$, which leads to the form $I_w^{\text{int}}(f) = \langle f'/(K_{\text{Sint}} - f') \rangle^{1/2}$ (shown in Fig. 1A). The observed broadening of the well line shape be-

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**FIG. 2.** The $^{71}$Ga OPNMR spectra (solid lines) of sample 10W at $T=0.46\text{K}$, for $0.267<\nu<1/3$ ($0^\circ<\theta<36.8^\circ$).

**FIG. 3.** The temperature dependence of the Knight shift (solid symbols) and the linewidth (open symbols) for several filling factors $0.267<\nu<1/3$ in sample 10W. Lines are to guide the eye.
time-averaged local polarization, which is equal to the total polarization \(P_{\text{total}}=0.828\) at \(\nu=0.275\) for our assumptions. At the other extreme \((\tau_j \rightarrow \infty)\), the motion is frozen out, and the single resonance is split into up and down lines, with areas proportional to \(p_+\) and \((1-p_+)\), respectively. Even within this simple model, the inhomogeneous breadth of the frozen line shape (owing to the quantum confinement) leads to a nontrivial evolution of the spectrum in the intermediate motion regime (for example, a given value of \(\tau_j\) might be simultaneously “fast” for nuclei at the edge of the well and “slow” for nuclei in the center of the well). In the intermediate motion regime, the FWHM of the \(w\) peak goes through a maximum when \(\tau_j=40\,\mu s\). Although varying the parameters \(p_+\) and \(K_{S\text{int}}\) (over the range relevant for samples 10W and 40W) affects the extreme value of the FWHM, the characteristic \(\tau_j\) remains \(\sim 40\,\mu s\).

On the basis of this simple model, the peaks in the FWHM at \(T_{\text{loc}} \approx 0.5\,K\) (Fig. 3) reflect the localization temperature of reversed spins, such that they fail to cover the sample uniformly over \(\sim 40\,\mu s\). The self-similar curves in Fig. 3 suggest that \(T_{\text{loc}}\) is not a strong function of the filling factor (or the density of reversed spins) for \(\nu < 1/3\). Below 0.5 K, the measured \(K_S(\nu=\frac{1}{3})\) increases toward \(K_S(\nu=\frac{1}{2})\), as seen in the model. However, even down to \(T=0.3\,K\), the spectra do not appear to match the frozen limit of our simulation. As \(\nu\) is varied below 1/3, the trends in the \(K_S\) and FWHM data (Fig. 3) continue smoothly through \(\nu=2/7\) without interruption. High-field magnetotransport measurements on samples taken from the same wafer as 10W show much more structure, with well-developed minima in \(\rho_{xx}\) at \(\nu=\frac{1}{5}, \frac{2}{5}, \frac{3}{5}\), and \(\frac{4}{5}\) at \(T = 0.3\,K\) [11,23].

Additional measurements of the linewidth for \(\nu > 1/3\) in sample 10W were consistent with the above picture. Measurements in sample 40W for \(\nu < 1/3\) were also in qualitative agreement, with one important quantitative difference: \(T_{\text{loc}}\) appeared to be shifted lower, so that only the high temperature side of the peak in the FWHM was observed down to \(T \approx 0.3\,K\). There was a similar sample variation in the saturation temperature of \(P(\frac{1}{2})\), with \(T_{\text{sat}}^{10W} \approx 0.77\,K\) and \(T_{\text{sat}}^{40W} \approx 0.46\,K\). The observed spectra contain more information than our simple simulation has revealed. A more sophisticated model might include: (i) a detailed structure for the reversed spin regions present below \(\nu=1/3\), (ii) the 2D dynamics of these reversed spins, and (iii) the effects of thermally excited spin flips, because \(T_{\text{sat}}\) is not that much greater than the \(T_{\text{loc}}\).

All of the results described thus far were acquired by applying a weak rf tipping pulse long after optical pumping to probe the equilibrium properties of the 2DES. Non-equilibrium properties of the electron spin system can be studied by varying these parameters at low temperatures, with a number of notable results at \(\nu=1/3\).

The rf tipping pulse for the NMR experiment is produced by a coil wrapped around the sample (Fig. 5C, in-

![Simulated NMR spectra simulated with the model described in the text. \(K_{S\text{int}}\) is set to 12 kHz for \(P=1\). The barrier is suppressed (\(a_0=0\)) for clarity.](image-url)
spin temperature on the rf pulse length (\(T = 0\)) for bars for function of dark time
axis) views of the grooved sapphire platform holding a sample circles 40W), and
bath (T) data for (S) FIG. 6. Spectra, which were acquired with sample 40W at
K

\(T\) sample 40W and (D) sample 10W. The intercept of the effective
S calibration curves based on the equilibrium
K

S(T) data for (A) sample 40W and (B) sample 10W. Error bars for \(K_S\) are shown. The dependence of the effective spin temperature on the rf pulse length (\(H_1 \approx 7\) Gauss) for (C) sample 40W and (D) sample 10W. The intercept of the straight line fit was constrained to be the lattice temperature: \(T = 0.31K\) (solid circles 10W and 40W), \(T = 0.42K\) (open circles 40W), and \(T = 0.44K\) (open circles 10W). The inset (C) shows the top (along \(\vec{z}'\)) and the front (along the rotation axis) views of the grooved sapphire platform holding a sample in a 5-turn rf coil.

The nonequilibrium spectra remain motionally narrowed, and appear to be indistinguishable from the corresponding equilibrium spectra measured at a higher lattice temperature. Thus, the electron spin system achieves internal equilibrium before our measurement, justifying our use of \(T_{spin}\) [10]. However, our measurement also shows that \(T_{spin}\) remains greater than \(T\) long after the rf pulse is turned off, which implies that the electron spin-lattice relaxation time \(\tau_{1s}\) is greater than 100 \(\mu\)s for \(T < 0.5\) K at \(\nu = \frac{1}{3}\).

The evolution of the spectra as the dark time \(\tau_D\) is increased provides an upper bound on \(\tau_{1s}\) (Fig. 6). The measured spectra are essentially independent of \(\tau_D\) after the first 0.5 s, which is consistent with the equilibration time of the the laser-heated sample with the helium bath at 0.45 K. Combining these results, we find 100 \(\mu\)s < \(\tau_{1s}\) < 500 ms for \(T < 0.5\) K at \(\nu = 1/3\). Although this value of \(\tau_{1s}\) is at least a factor of 1000 longer than recent measurements of the transverse relaxation time \(\tau^*_2\) in bulk GaAs [24], it is consistent with a previous theoretical prediction [23] that had assumed conditions similar to those in our experiment.
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