Dispersion line shape in the vicinity of the $\nu = 1$ quantum Hall state: Coexistence of Knight shifted and unshifted RDNMR responses

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The frequency splitting between the dip and the peak of the resistively detected nuclear magnetic resonance (RDNMR) dispersive line shape (DLS) has been measured in the quantum Hall effect regime as a function of filling factor, carrier density and nuclear isotope. The splitting increases as the filling factor tends to $\nu = 1$ and is proportional to the hyperfine coupling, similar to the usual Knight shift versus $\nu$ dependence. The peak frequency shifts linearly with magnetic field throughout the studied filling range and matches the unshifted substrate signal, detected by classical NMR. Thus, the evolution of the splitting is entirely due to the changing Knight shift of the dip feature. The nuclear spin relaxation time, $T_1$, is extremely long (hours) at precisely the peak frequency. These results are consistent with the local formation of a $\nu = 2$ phase due to the existence of spin singlet $D^{-}$ complexes.

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Resistively detected nuclear magnetic resonance (RDNMR) is well-established technique to probe the interaction between nuclear and electronic spin systems via the contact hyperfine interaction. For the investigation of two-dimensional electron gases (2DEGs), RDNMR has significantly increased sensitivity with respect to classical NMR, as it only probes nuclei which have significant overlap with the electronic wave function. Historically, RDNMR was developed to demonstrate the role played by nuclear spins in the formation of a huge longitudinal resistance spike at filling factor $\nu = 2/3$ and has since proved to be a formidable tool to investigate quantum Hall physics.

In particular, attention has been focussed on the $\nu = 1$ QH state following the observation of an unexpected dispersive line shape (DLS). A strong coupling of nuclear spins to low-energy (gapless) excitations of the many-body quantum Hall ground state in the vicinity of $\nu = 1$ was invoked to explain the anomalous resistance spike. Notably, to explain the unusually short nuclear spin relaxation times $T_1$ observed, it was suggested that a coupling occurs with the Goldstone modes of a skyrmion crystal. However, in later investigations the DLS was detected at $\nu = 1$ under unfavorable conditions for the formation of skyrmions. It is now evident that skyrmions alone cannot account for the DLS and indeed a coherent description of the origin of the dispersive line shape is still lacking. Thermal effects have been put forward by Tracy et al. to explain the coincidence between the DLS shape inversion and the $dR_{xx}/dT$ sign change. Separate mechanisms for the dip and peak components have been proposed, since they show different $T_1$ behaviors versus filling factor and respond differently to dc-current. Recently, the frequency splitting between the dip and the peak of the DLS, at a constant filling factor, was shown to increase linearly versus the magnetic field in the $5 - 16$ T range.

Here we report on the systematic investigation of dispersive line shapes around $\nu = 1$ as a function of filling factor, electron density, and nuclear isotope for three different samples at mK temperatures. The frequency splitting between the dip and peak resonances in the magnetoresistance is shown to increase as $\nu \rightarrow 1$ and is proportional to the carrier concentration and the hyperfine interaction. More precisely, we demonstrate the peak frequency shifts linearly with magnetic field and coincides with the response of nuclei which are not coupled to a polarized electronic system, obtained by classical NMR. In contrast, the dip feature shows significant deviation from linearity reflecting the variation of the polarization of the electronic system (Knight shift). Extremely long nuclear spin relaxation times ($T_1 \sim 1$ hr) are also measured at the peak frequency consistent with the local formation of a $\nu = 2$ (unpolarized) phase due to the existence of spin singlet $D^{-}$ complexes.

RDNMR has been performed on three GaAs/AlGaAs heterojunctions patterned into Hall bars. The carrier densities and mobilities are $1.56$, $1.63$ and $2.13 \times 10^{11}$ cm$^{-2}$ and $5.8$, $1.46$ and $0.82 \times 10^{6}$ cm$^{-2}$V$^{-1}$s$^{-1}$ for samples 1 (NRC 1707), 2 (NRC V0050) and 3 (NU 2077) respectively. Each device was placed in the mixing chamber of a dilution fridge with a base temperature of $T = 10$ mK. The radio-frequency (RF) field was applied by means of a copper coil wound around the sample and connected to an RF-synthesizer by a rigid coaxial cable. The typical RF power used was around $\sim 5$ dBm, which gives a significant RDNMR signal while avoiding excessive heating (the electron temperature is estimated...
Resonant frequency is given by $\Delta f_{DLS}$ is the frequency splitting between the resistance dip and peak. Successive RDNMR spectra have been recorded at several magnetic fields under the same conditions for all three samples. Fig. 1(a) shows a typical RDNMR dispersive line shape measured by RD-NMR at $\nu = 0.83$. $\Delta f_{DLS}$ is the nuclear gyromagnetic ratio and $B_e$, the effective field seen by the nucleus due to the contact hyperfine interaction. A close inspection reveals that the peak response shifts linearly with magnetic field suggesting that it is not influenced by the polarization of the electronic system. Assuming that the influence of $B_e$ is negligible for the peak response, the Larmor frequency can be subtracted from the RDNMR data, by fitting a linear dependence to the peak resonance frequency versus $B_e$. The frequency positions of the DLS peak and dip from which the Larmor frequency has been subtracted are plotted in Fig. 1(b) as a function of the filling factor for sample $\sharp g2$. We observe that the peak frequency remains constant throughout the $0.8 - 1.2$ filling factor range. In contrast, the dip frequency is shifted to lower frequencies as $\nu$ tends to one from either above or below. We should stress that the detection of the RDNMR deep into the quantized regime (QHE regime of dissipationless magnetoresistance, see Fig. 1(c)) was made possible by applying increasing dc currents (up to 5 $\mu$A), as described by Dean et al. The closed diamonds in Fig. 1(b) represent data obtained without dc current far from $\nu = 1$.

The very different dependence of the peak and dip frequencies upon the filling factor suggests they originate from different NMR responses. In order to confirm this point, classical NMR has been performed on sample $\sharp 2$. Figure 1(d) shows the Fourier transform of the free induction decay signal of $^{71}$Ga nuclei obtained by classical NMR and RDNMR performed at exactly the same magnetic field. The peak of the NMR signal indicates the resonant frequency of the majority nuclei in the sample, i.e. those in the undoped barriers and substrate, which are not coupled to conduction electrons. In contrast, the RDNMR signal is only sensitive to the nuclei which are in contact with the wave function of the 2DEG. These nuclei will feel the spin polarization of the 2DEG and their NMR frequency should be Knight shifted accordingly. The peak position of the RDNMR line for the RF upswEEP occurs at a higher frequency than for the RF downswEEP, but this discrepancy reduces as the RF sweep rate becomes slower. The inset of Fig. 2, which plots the DLS peak position for up and down sweeps as a function of the RF sweep rate, suggests the peak positions will converge to the reference NMR frequency for an experimentally unrealistic sweep rate of 10 mHz/s. Thus, the peak response originates from nuclei which are in contact with the 2DEG, but nevertheless see zero electronic polarization ($B_e = 0$) over a wide range of filling factors.

The frequency splitting of the dispersive line, $\Delta f_{DLS}$, i.e. the absolute difference between the dip and peak frequencies; plotted in Fig. 1(a); was found to be perfectly reproducible for a given constant RF sweep rate $df/dt = 160$ Hz/s. The DLS, which is characterized by a resistance dip followed by a peak at a higher radio frequency, is observed on both sides of $\nu = 1$ for all three samples. Successive RDNMR spectra have been recorded at several magnetic fields under the same temperature and RF sweep rate conditions. The nuclear resonant frequency is given by $f = \frac{\gamma}{2}(B + B_e)$, where $\gamma$ is the nuclear gyromagnetic ratio and $B_e$, the effective field seen by the nucleus due to the contact hyperfine interaction. A close inspection reveals that the peak response shifts linearly with magnetic field suggesting that it is not influenced by the polarization of the electronic system. Assuming that the influence of $B_e$ is negligible for the peak response, the Larmor frequency can be subtracted from the RDNMR data, by fitting a linear dependence to the peak resonance frequency versus $B_e$. The frequency positions of the DLS peak and dip from which the Larmor frequency has been subtracted are plotted in Fig. 1(b) as a function of the filling factor for sample $\sharp g2$. We observe that the peak frequency remains constant throughout the $0.8 - 1.2$ filling factor range. In contrast, the dip frequency is shifted to lower frequencies as $\nu$ tends to one from either above or below. We should stress that the detection of the RDNMR deep into the quantized regime (QHE regime of dissipationless magnetoresistance, see Fig. 1(c)) was made possible by applying increasing dc currents (up to 5 $\mu$A), as described by Dean et al. The closed diamonds in Fig. 1(b) represent data obtained without dc current far from $\nu = 1$.

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Fourier transform of the free induction decay signal (filled circles, right axis) of $^{71}$Ga measured at $B = 10.8$ T on sample $\gamma 3$. The vertical dashed line shows the NMR substrate frequency and arrows stand for the RF sweep directions. In inset, DLS peak frequencies for RF-upsweeps (disks) and downsweeps (circles) vs RF sweep rate. The dashed line shows the classical NMR signal frequency. Dotted lines are a guide to the eye.

larger for higher density 2DEGs, which is in qualitative agreement with the above picture. A quantitative comparison cannot be performed as the extent of the 2DEG electronic wave functions are not precisely known.

For sample $\gamma 3$ we have also measured the frequency splitting as a function of the 2DEG density $n_s$, which was modified by successive in situ illuminations with an infrared LED, using the persistent photo-conductivity effect. Fig. (c) shows the frequency splitting multiplied by the density ratio $n_s^{\text{dark}}/n_s^{\text{light}}$ for densities 2.13, 2.66, 2.90 and $3.26 \times 10^{11}$ cm$^{-2}$ (squares, up and down triangles, and diamonds resp.). The superimposition of all the data sets confirms that as for a Knight shift, the DLS frequency splitting is proportional to the electron density. Finally, the ratio of the frequency splitting of the $^{75}$As and $^{69}$Ga isotopes in respect of $\Delta f_{\text{DLS}}$ for $^{71}$Ga are plotted in Fig. (f), for the high magnetic field flank of $\nu = 1$ (sample $\gamma 3$). We see that for each nuclear species the ratio is almost constant over the measured field range, i.e. 0.899(0.773) for $^{75}$As($^{69}$Ga), in agreement within 5% with the expected $\gamma |\Psi_0|^2$ values (0.948 and 0.787 resp.). To summarize, Figs. (d-f) demonstrate unambiguously that the DLS frequency splitting $\Delta f_{\text{DLS}}$ depends on the filling factor $\nu$, the electron density $n_s$ and the hyperfine interaction $A$, in exactly the same way as the Knight shift. Note that the measured frequency splittings $\Delta f_{\text{DLS}}$ range within 5 – 30 kHz, which agree well the expected hyperfine splittings $A$ in 2DEGs, scaled by the $n_s/w$ factor.

The fact that the frequency position of the DLS peak remains constant versus filling factor around $\nu = 1$ and matches the classical NMR reference frequency demonstrates unequivocally that it reflects the response of nuclei which although in contact with the 2DEG feel no nett polarization of the electronic system. To further probe this state, we have measured the transient resistance when the RF is switched on and off, throughout the DLS resonance (see Ref.2 for details of this technique). The resistance transient during an RF on/off sequence is plotted in Fig. (d). The frequency used here is $f = 57.105$ MHz, which corresponds to the resonant frequency of the DLS peak for $B = 7.87$ T (Fig. (a)). When the RF is switched on, the resistance first increases rapidly and then drifts slowly without ever reaching a steady state on an experimentally viable timescale (several hours). This behavior is distinctive of frequencies close to the peak feature. For other frequencies and under the same experimental conditions, the resistance always saturates. Similar observations were reported by Kodera et al., who found long relaxation time scales for the DLS peak.

The resistance relaxation, after the RF is switched off, is well fitted by a double exponential (symbols in Fig. (c)). Both slow and fast nuclear spin relaxation times extracted from these fits are plotted in Fig. (b). It is evident that the slow $T_1$ shows a sharp maximum at $f = 57.105$ MHz which is exactly the DLS peak position indicated on the figure by the vertical dashed line. The involvement of an extremely slow process is further confirmed by the dispersive line recorded at a very low RF-

![FIG. 2. RDNMR dispersive lines (upper curves, left axis) and Fourier transform of the free induction decay signal (filled circles, right axis) of $^{71}$Ga measured at $B = 10.8$ T on sample $\gamma 3$. The vertical dashed line shows the NMR substrate frequency and arrows stand for the RF sweep directions. In inset, DLS peak frequencies for RF-upsweeps (disks) and downsweeps (circles) vs RF sweep rate. The dashed line shows the classical NMR signal frequency. Dotted lines are a guide to the eye.](attachment:image.png)

![FIG. 3. (a) DLS at $B = 7.87$ T for RF up/downsweeps (solid lines) and under static conditions (dots). (b) Slow and fast $T_1$ vs radio-frequency $f$ (filled diamonds and circles resp.). The vertical dashed line indicates the peak resonance position at $f = 57.105$ MHz. (c) Time dependence of $R_{xx}$ for an RF on/off sequence at the peak frequency $f = 57.105$ MHz. Double exponential relaxation fit (circles). (d) DLS measured at $B = 7.82$ T with a slow RF-sweep rate of 8 Hz/s (solid line). The top axis indicates the elapsed time in hours. For comparison, the DLS measured with a 100 times faster sweep rate (800 Hz/s) is plotted (dashed line). Double exponential fit of the slow trace relaxation (circles).](attachment:image.png)
sweep rate (8 Hz/s) plotted in Fig. 3d). The total scan duration is 4 hours (top axis). Assuming that the return to equilibrium is mainly limited by $T_1$, the double exponential relaxation rate above the peak frequency, leads to a characteristic time of the order of 1 hr. In addition, it is interesting to note that the DLS with an RF-sweep rate of 800 Hz/s (dashed line) has a frequency splitting $\Delta f_{DLS}$ almost equal to the DLS recorded with a 100 times slower sweep rate. This justifies a posteriori the choice of an intermediate sweep rate of 160 Hz/s for the measurement of the frequency splittings $\Delta f_{DLS}$ presented in Fig. 1.

Thus, the dispersive line shape should be understood as the NMR responses of nuclei interacting with two electron subsystems of the 2DEG. The dip corresponds to the Knight shifted resonance of nuclei coupled to partially spin polarized electrons. The observed shift is peaked at filling factor $\nu = 1$ for which the polarization of the 2DEG is a maximum. The fast nuclear spin relaxation times observed imply that the electron system has low energy spin excitations which favors nuclear spin relaxation via the flip-flop process in which energy is conserved. The Korringa law predicts that $T_1^{-1} \propto D_{\nu}(E_f)D_{\nu}(E_f)$ is proportional to the density of spin up and spin down states at the Fermi energy which precludes the presence of a significant spin gap in the electronic system. Within a single electron picture, on both sides of filling factor $\nu = 1$, the Fermi energy lies in a partially occupied spin Landau level and an electronic spin excitation requires an energy $\mu_B B$ which is orders of magnitude larger than the nuclear spin flip energy. However, the physics in the vicinity of filling factor $\nu = 1$ is known to be extremely rich with the possibility to form spin reversed ground states i.e. spin textures such as skyrmions which are thought to have considerably smaller gaps for electronic spin flip excitations. The single electron picture also predicts that the 2DEG should be completely polarized for filling factors below $\nu = 1$. In reality many body physics dominates and it is experimentally well established that the electronic polarization collapses rapidly on either side of $\nu = 1$. On the other hand, the peak feature of the DLS is the unequivocal signature of a zero Knight shift of nuclei coupled to an unpolarized 2DEG. For the range of filling factors investigated this is simply not possible for the case of electrons in uniformly occupied Landau levels. Thus, the 2DEG has to form domains with polarized and unpolarized regions. In addition, these domains have to be localized on the time scale of the NMR measurements to prevent the nuclei simply feeling the average spin polarization of the 2DEG. While it would not be surprising that the 2DEG could spontaneously break symmetry to form a novel ground state with the presence of domains, there is actually a much simpler explanation. Recent inelastic light scattering experiments have demonstrated the preponderant role played by $D^-$ complexes in quantum Hall states and the existence of a depolarized electron subsystem at $\nu = 1$ in particular. These complexes are formed by two electrons of opposite spins bound to a positively charged donor impurity located in the barrier. The electrons remain in the lowest Landau level of the 2DEG and this spin singlet state can be thought of as a local region in which the filling factor is $\nu = 2$. The $D^-$ electrons are weakly bound and extend over many nuclei with a Bohr radius $ab \approx 400 A$. This should be compared with the magnetic length $\ell_B \approx \sqrt{\hbar/eB} \approx 230 A$ at $B = 8 T$. Other electrons in the 2DEG are repelled from $D^-$ both by the Pauli principle and the Coulomb repulsion. At $\nu = 2$ the Fermi energy lies in the cyclotron gap so that Korringa predicts a very long $T_1$ since nuclear spin relaxation can only proceed via slow processes such as nuclear spin diffusion. We note that very recent experimental RDNMR results near filling factor $\nu = 2$ have revealed a reduced Knight shift, as well as long relaxation times and were interpreted as the formation of an electron solid phase.

To conclude, the splitting of the peak and dip features of the dispersive line shape RDNMR signal in the vicinity of $\nu = 1$ can be used to determine the Knight shift and thus the spin polarization of the 2DEG. Classical NMR confirms that the peak feature, which originates from nuclei in contact with an unpolarized phase of the 2DEG, provides a convenient relative zero Knight shift reference signal.

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