Resistively Detected Nuclear Magnetic Resonance in the Quantum Hall Regime: Possible Evidence for a Skyrme Crystal

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Resistively detected nuclear magnetic resonance measurements have been performed on a high mobility heterostructure in the quantum Hall regime. At millikelvin temperatures the nuclear resonances are observed in the vicinity of various integer and fractional filling factors without previous dynamic nuclear polarization. Near $\nu = 1$, the observed large enhancement of the resonance amplitude accompanied by a reduction of T_1 strongly suggests a greatly increased coupling between the electronic and nuclear spin systems. This is consistent with the proposed coupling of the nuclear spin system to the Goldstone mode of the Skyrme crystal.

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The coupling of electrons in a two-dimensional electron gas (2–DEG) and nuclei via the hyperfine interaction has previously been studied using electron spin resonance (ESR) to measure the Overhauser shift [1,2] or inversely using nuclear magnetic resonance (NMR) to determine the Knight shift [3,4]. Recently, Kronmüller and co-workers [5,6] reported an anomalous resistance maximum at filling factor $\nu = 2/3$. A strong coupling between the electrons and nuclei was inferred from the large time constants observed. This was confirmed by resistively detected NMR which could be measured without recourse to dynamic pumping of the nuclear spin system at $T = 0.35$ K. Resistively detected NMR has previously been observed but only under special conditions in which the nuclear spin system is first dynamically polarized [7,8].

Here we show that at sufficiently low temperature the nuclear resonances can be observed without recourse to dynamic nuclear spin polarization for a wide range of odd, even, and fractional filling factors in standard quantum Hall samples which do not show an anomalous peak at $\nu = 2/3$. The coupling between the electrons and the nuclear system occurs via the hyperfine interaction which modifies the electronic Zeeman energy. Close to filling factor $\nu = 1$ an anomalous line shape together with a large enhancement of the amplitude of the NMR resonance is observed. The enhanced coupling is in agreement with specific heat capacity measurements [9,10] and can be interpreted as evidence for coupling of the nuclei to the low energy modes of the Skyrme crystal which is expected to form in the vicinity of $\nu = 1$ [11,12].

The measurements have been performed on a high mobility GaAs/AlGaAs heterojunction ($\mu = 5 \times 10^6$ cm^2 V^{-1} s^{-1}) with a carrier density of $n = 1.6 \times 10^{11}$ cm^{-2}. A 250 μm wide Hall bar was patterned with 750 μm between the voltage contacts. The longitudinal resistance R_{xx} is plotted as a function of the magnetic field at $T = 50$ mK in Fig. 1a. The resistance is measured under quasi-dc conditions ($I = 100$ nA, $f = 10.7$ Hz) using a classical lock-in technique. For the NMR measurements the sample is mounted in the mixing chamber of a dilution refrigerator. Low temperatures are required in order to obtain a significant nuclear polarization. At $T = 50$ mK and $B = 10$ T we estimate that the typical thermal nuclear polarization is around 10%. In the inset of Fig. 1a, a schematic of the experimental setup is shown. The static magnetic field produced by a 15-T superconducting magnet is applied normal to the 2-DEG. The radio frequency (rf) field is generated by a single turn coil wound around the sample. The typical rf voltage injected into the coil is 50 mV, and we estimate the rf field H_1 to be of the order of 1 μT. The nonresonant heating of the electron system due to the application of the rf field leads to an effective electronic temperature which is typically 50-100 mK above base temperature [13].

Resistively detected NMR is performed by sweeping the radio frequency through the nuclear resonance frequency while the static magnetic field is maintained at a fixed value. A typical resonance line is shown in Fig. 1b which represents the longitudinal resistance R_{xx} versus the frequency of the applied rf signal measured at $B = 3.65$ T for the ^{75}As nuclei. The solid line is an upsweep obtained with an rf-sweep rate of 4 kHz/s. Initially, the longitudinal resistance remains constant, and when the resonance frequency is reached, R_{xx} exhibits a sudden decrease followed by an exponential relaxation back to its equilibrium value. The exponential relaxation is governed by the nuclear spin

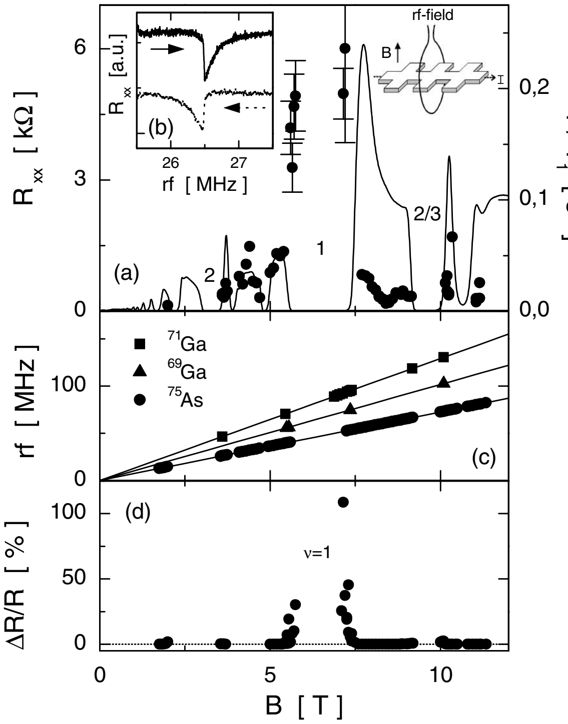

FIG. 1. (a) Longitudinal resistance R_{xx} (solid line) and the nuclear spin relaxation rate $1/T_1$ (•) as a function of the magnetic field. The experimental configuration is represented schematically in the inset. (b) A typical ^{75}As resonance line obtained at $B = 3.65$ T. The arrows indicate the rf sweep direction (curves offset vertically for clarity). (c) Dependence of the resonance frequency on the magnetic field for the three isotopes. (d) Magnetic field dependence of the resistance amplitude $\Delta R_{xx}/R_{xx}$ for the ^{75}As nuclei.

relaxation time T_1. A similar behavior is observed when sweeping through the resonance from above (dashed line Fig. 1b). Typical values of T_1 determined from the exponential relaxation are ~30 s.

Resonance lines have been observed for all three isotopes ^{75}As, ^{69}Ga, and ^{71}Ga. Here we focus on ^{75}As since its resonance line exhibits the largest amplitude due to its abundance. As expected no resonance is found for ^{27}Al since the hyperfine coupling between electronic spins in the 2-DEG and nuclear spins in the barrier is negligible. Figure 1c shows the linear magnetic field dependence of the resonance frequency of each nucleus. The slope gives the gyromagnetic ratio γ of the corresponding isotope. The values found are in agreement with those reported in the literature [14]: $\gamma_{^{69}\mathrm{Ga}} = 63.96$ MHz/T, $\gamma_{^{71}\mathrm{Ga}} = 81.24$ MHz/T and $\gamma_{^{75}\mathrm{As}} = 45.61$ MHz/T. From Fig. 1 we see that the nuclear resonances have been detected for odd ($\nu = 1, 3$), even ($\nu = 2, 4$), and fractional filling factors ($\nu = 3/5, 2/3, 4/3, 5/3$). In other words, the resistively detected NMR is observed over a large magnetic field range. The commonly measured resonance amplitudes $\Delta R_{xx}/R_{xx}$ are of the order of 0.5% for the majority of the filling factors (Fig. 1d). However, in the vicinity of $\nu = 1$ the observed amplitude of the resonance is enhanced by almost 2 orders of magnitude.

Reducing the rf sweep rate leads to a narrower line shape and in some cases allows one to resolve the quadrupole splitting. Figure 2 shows a ^{75}As resonance obtained at the same field as in Fig. 1b but at a slower sweep rate of 0.09 kHz/s. Under these conditions three resonances are observed separated by $\Delta\mathrm{rf} = 27$ kHz. This is the signature of the quadrupole splitting arising from the interaction of the nuclear electric quadrupole moment with an electric field gradient [15–17]. The quadrupole splitting Δrf measured here is in agreement with the previously reported values in GaAs [18].

The electronic spin ($\mathbf{S}$) and a nuclear spin ($\mathbf{I}$) couple via the hyperfine interaction $A\mathbf{I} \cdot \mathbf{S}$, where the hyperfine coupling constant A is proportional to the electronic density at the nuclear site. In the presence of an external magnetic field B and a nuclear spin polarization $\langle I_z \rangle$, the electronic Zeeman energy is given by

$$E_z = g^* \mu_B B S_z + A\langle I_z \rangle S_z, \tag{1}$$

where g^* is the effective electronic g factor. It is possible to define $B_N = A\langle I_z \rangle / g^* \mu_B$, the nuclear magnetic field experienced by the electronic spins, which is at the origin of the Overhauser shift. It is important to note that B_N arises as a result of the contact hyperfine interaction and has no influence on the orbital motion of the electrons so that the filling factor remains unchanged. The maximum nuclear field in GaAs can be as high as $B_N = 5.3$ T when all nuclei species are fully polarized [19]. Under thermal equilibrium, at $B = 10$ T and $T = 50$ mK, this nuclear field is around 0.1 T for a given isotope. Because of the negative sign of the electronic g factor in GaAs $g^* = -0.44$, a net nuclear polarization which is parallel to the external magnetic field reduces the electronic Zeeman gap (i.e., $B_N < 0$ under normal equilibrium conditions). By applying a radio frequency field under resonant conditions, it is possible to depolarize the nuclear spins (reducing the amplitude of B_N) and as a consequence to increase the electronic Zeeman gap.

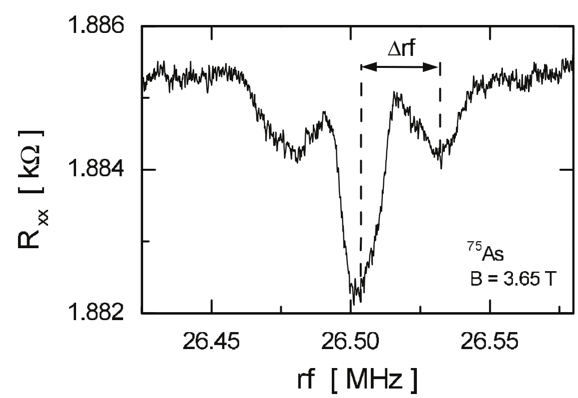

FIG. 2. ^{75}As resonance line exhibiting a quadrupole splitting at $B = 3.65$ T.

It therefore seems natural that nuclear spins couple to the resistance of the sample via the hyperfine interaction which modifies the electronic Zeeman energy. It is well established that the longitudinal resistance R_{xx} depends on the energy gap in the density of states. In the thermally activated regime $R_{xx} \propto \exp(-\Delta/2kT)$ or from the Ando [20] formula $R_{xx} \propto x/\sinh(x)$ where the term $x = 2\pi^2 k_B T/\Delta$ effectively depends on the energy gap Δ. Sweeping the rf through resonance reduces $\langle I_z \rangle$ and consequently increases the electronic Zeeman energy. For the case of odd filling factors, R_{xx} is sensitive to B_N since the gap is simply equal to the Zeeman gap, $\Delta = g^* \mu_B (B + B_N) + E_{\text{exch}}$ where E_{exch} is the exchange energy which results from the electron-electron interactions. For even filling factors the effective gap is equal to $\Delta = \hbar\omega_c - E_z$ where $\hbar\omega_c$ is the cyclotron energy. Finally, it is experimentally well established [21,22] and theoretically understood [23,24] that the gap for fractional filling factors can also depend on the Zeeman energy.

When saturating the nuclear spin system, i.e., $\langle I_z \rangle \to 0$, the measured resonance amplitude is around $\Delta R_{xx}/R_{xx} \approx 0.5\%$. The corresponding variation of the Zeeman energy is $\Delta E_z = g^* \mu_B B_N$, i.e., $\Delta E_z \approx +2.55\ \mu$eV, for the typical thermal equilibrium value $B_N = 0.1$ T. This model can be validated by the comparison with experiments in which the Zeeman energy is modified by rotating the sample in the external magnetic field. The change in Zeeman energy that corresponds to a tilt angle θ away from the normal magnetic field is $\Delta E_z = g^* \mu_B B_\perp (\frac{1}{\cos\theta} - 1)$. From such measurements performed on the same sample (not shown), we estimate that for a comparable Zeeman energy enhancement ΔE_z, the corresponding resistance amplitudes are in the range 0.6%–1% which is of the same order as observed for the NMR resonances.

Either side of filling factor $\nu = 1$ an anomalous line shape for the NMR resonance is observed as shown in Fig. 3b. This has been detected for the ^{75}As nuclei at $B = 7.4$ T corresponding to $\nu = 0.89$. We note that the line shape is similar to a traditional NMR dispersion line and has a huge amplitude which can be as large as 100%, i.e., almost 2 orders of magnitude larger than at any other filling factor (see Fig. 1d). The response of the system under static conditions has also been measured. The resistance R_{xx} as a function of time is shown in Fig. 3a. At a certain time (rf on) the desired rf close to the nuclear resonance is applied which induces a variation of the resistance until the equilibrium of the nuclear spin system is achieved. When the steady state is reached, the rf field is switched off (rf off) and the resistance decays exponentially to the unperturbed value. We see that the saturation values ΔR_{xx} (open circles in Fig. 3b) are superimposed on the dynamic curves obtained at an rf sweep rate of 4 kHz/s. The system is therefore under equilibrium conditions, indicating that the nuclear spin relaxation time is short. From the relaxation (observed after rf off) $1/T_1$ can be measured and is shown in Fig. 1a. The evolution of $1/T_1$ clearly

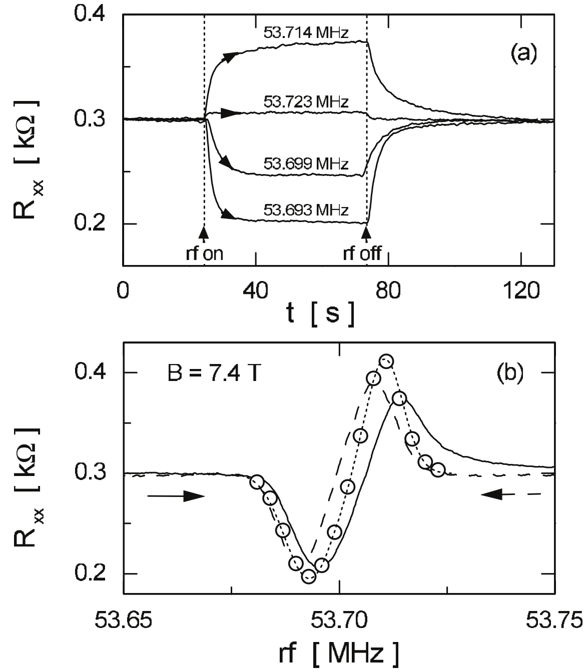

FIG. 3. (a) Time dependence of the longitudinal resistance R_{xx} under static radio frequency and fixed magnetic field. (b) Resonance lines obtained under dynamic conditions (solid and dashed lines). The open circles are the saturation values obtained under steady-state conditions. The dotted line is a guide for the eye.

follows the expected Korringa behavior ($1/T_1$ is proportional to the density of states at the Fermi level and hence proportional to the sample resistance) except in the region of filling factor 1 where a large enhancement of the relaxation rate is observed. The deduced relaxation rates near $\nu = 1$ are probably limited by the time constant of the lock-in amplifier used to measure the resistance so that $T_1 \lesssim 3$ s in agreement with the recent values found by Hashimoto [25].

An enhanced coupling between the electrons and the nuclear spins due to the formation of spin textures (Skyrmions) has recently been invoked [9,10] in order to explain the anomalously large heat capacity near $\nu = 1$. Theoretically, it has been shown that a coupling to a gapless spin wave excitation of the Skyrme crystal (Goldstone mode) could be at the origin of the dramatic enhancement of the nuclear spin relaxation rate $1/T_1$ [11,12]. In our sample close to $\nu = 1$ the dimensionless Zeeman energy $\eta = g^* \mu_B B/(e^2/\epsilon \ell_B) \sim 0.014$ is favorable for the formation of Skyrmions [26] in the ground state [$\ell_B = (\hbar/eB)^{1/2}$ is the magnetic length]. We therefore suggest that the coupling of the nuclear spins to the gapless spin-wave excitations of the Skyrme crystal is responsible for the observed huge amplitude and anomalous line shape of the resistively detected NMR around $\nu = 1$.

A crucial test for this interpretation is the behavior of the NMR resonance as the sample is tilted to modify η.

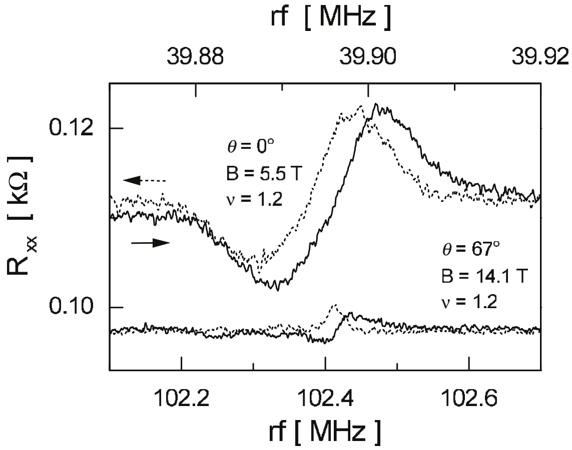

FIG. 4. ^{75}As resonance lines obtained at filling factor $\nu = 1.2$ under perpendicular and tilted fields.

The filling factor depends only on the perpendicular component of magnetic field while the Zeeman energy depends on the total magnetic field. Figure 4 shows two resonance lines detected at the same filling factor $\nu = 1.2$ but at total magnetic fields of $B = 5.5$ T (perpendicular) and $B = 14.1$ T (tilted). In the absence of Skyrmions, for a given filling factor, we would expect the amplitude of the resonance to increase with increasing total magnetic field, due to the increased nuclear spin polarization. In contrast, we observe that the amplitude of the resonance line for the tilted case is about 5 times smaller than under normal field. At $B = 14.1$ T the ratio ($\eta \sim 0.022$) is close to the limit where the creation of Skyrmions is no longer energetically favorable [26]. The reduction of the nuclear resonance amplitude at high field is therefore consistent with the coupling of the nuclear spins to the low-energy excitations of the Skyrme crystal. However, further measurements at higher magnetic fields are required in order to achieve the limit $\eta \gg 0.02$ where Skyrmion formation can be excluded and in principle a normal line shape should be recovered.

A further test for the model is the case of filling factor $\nu = 3$ for which the formation of Skyrmions is not expected theoretically [27,28] or observed experimentally [29]. Rotating the sample in magnetic field we can place $\nu = 3$ at a total magnetic field ~5 T close to that for filling factor $\nu = 1$ in the perpendicular configuration. This allows one to study $\nu = 3$ under identical conditions to those in which an anomalous line shape and huge amplitude are observed for $\nu = 1$. We have carefully investigated the ^{75}As NMR resonance in the vicinity of $\nu = 3$ and find no evidence for the existence of an anomalous line shape or an enhanced amplitude, in agreement with the predicted absence of Skyrmions at filling factor $\nu = 3$.

Clearly the ensemble of the data presented here supports the hypothesis that the anomalous line shape and large resonance amplitude observed near $\nu = 1$ are closely linked to the presence of Skyrmions in the ground state. Our observations are consistent with the theoretically proposed coupling of the nuclear spins to the Goldstone mode of the Skyrme crystal [11,12].

Note added.—After the submission of this manuscript a coupling of the nuclei to the Skyrme crystal has also been proposed by Smet and co-workers [30].

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