Evidence for Skyrmion Crystallization from NMR Relaxation Experiments

G. Gervais\textsuperscript{1,2}, H.L. Stormer\textsuperscript{1,3}, D.C. Tsui\textsuperscript{4}, P.L. Kuhns\textsuperscript{2}, W.G. Moulton\textsuperscript{2}, A.P. Reyes\textsuperscript{2}, L.N. Pfeiffer\textsuperscript{3}, K. W. Baldwin\textsuperscript{3}, and K.W. West\textsuperscript{3}

\textsuperscript{1}Department of Physics and Department of Applied Physics, Columbia University, New York, NY 10027 USA
\textsuperscript{2}National High Magnetic Field Laboratory, Tallahassee, FL 32306, USA
\textsuperscript{3}Bell Laboratories, Lucent Technology, Murray Hill, NJ 07974 USA
\textsuperscript{4}Department of Electrical Engineering, Princeton University Princeton, NJ 08544 USA

A resistively detected NMR technique was used to probe the two-dimensional electron gas in a GaAs/AlGaAs quantum well. The spin-lattice relaxation rate ($1/T_1$) was extracted at near complete filling of the first Landau level by electrons. The nuclear spin of $^{75}$As is found to relax much more efficiently with $T \to 0$ and when a well developed quantum Hall state with $R_{xx} \approx 0$ occurs. The data show a remarkable correlation between the nuclear spin relaxation and localization. This suggests that the magnetic ground state near complete filling of the first Landau level may contain a lattice of topological spin texture, i.e. a Skyrmion crystal.

PACS numbers: 73.43.-f, 73.43.Fj, 75.30.Ds

In his seminal work on nuclear matter more than forty years ago Skyrme showed that baryons emerge mathematically as a static solution of a meson field described by the so-called Skyrme Lagrangian \cite{1}. His work provided the foundation for the quantum theory of solitons, and more recently found an interesting and \textit{a priori} surprising connection to the physics of electrons confined to a two-dimensional plane. In the presence of a strong perpendicular magnetic field, the orbital motion of these electrons is quantized into discrete Landau levels. When only the lowest of such level is almost completely occupied, the elementary excitations of the system become large topologically stable spin texture known as Skyrmions \cite{2}. It was further proposed that at $T=0$ Skyrmions would localize on a square lattice \cite{3}. This ground state represents a new type of magnetic ordering which possesses long-range orientation and positional order. This is the solid-state analogue of the Skyrmion crystal state which is used to describe dense nuclear matter using Skyrme’s topological excitation model \cite{4}. In this work, we present an extensive study of nuclear magnetic resonance (NMR) spin-lattice relaxation rate in the first Landau level of an extremely high-quality GaAs/AlGaAs sample. We find strong and enhanced relaxation in the limit of $T \to 0$ and $R_{xx} \to 0$ where localization of electronic states occurs. This is consistent with previous measurements of the heat capacity in multiple quantum wells at very low temperatures \cite{5,6}, and with the predictions of a magnetic ground state containing a Skyrmion crystal \cite{7}.

Several theoretical publications \cite{2,3,7,8,9,10,11,12} have pointed toward the existence of Skyrmions in a two-dimensional electron gas (2DEG). At filling factor $\nu = 1$, where $\nu$ is defined by the ratio of the electronic density $n$ to the magnetic flux density, $\nu = \frac{\Phi}{\Phi_0} = \frac{n eB}{\Phi_0}$, the quantized Hall state is ferromagnetic. For sufficiently small Zeeman-to-Coulomb energy ratio $\eta = E_z/E_c = \frac{g^2 \mu_B B}{\sqrt{\hbar/eB}}$, where $g^*$ is the electronic g-factor and $I_B = \sqrt{\hbar/eB}$ is the magnetic length, Sondhi \textit{et al.} showed that the low-lying excitations are not single spin-flips, but rather a smooth distortion of the spin field in which several spins (4-30) participate \cite{2}. These Skyrmions are topologically stable, charged $\pm e$, and gapped excitations which are the result of an energy tradeoff where a higher Zeeman cost is paid for the profit of lowering the exchange energy between neighboring spins. While several theoretical proposals suggest a lattice state of Skyrmions \cite{8,10,11} it is theoretically debated \cite{12} whether this is the case or whether a ‘liquid’ state prevails.

Previous measurements of the electronic spin polarization by NMR around $\nu = 1$ by Barrett \textit{et al.} showed strong evidence for a finite density of Skyrmions at temperature $T \sim 1K$ \cite{13,14}. Subsequently, Schmeller \textit{et al.} \cite{15} used tilted-transport measurements to show that as many as seven electron spins participate in the spin excitations at $\nu = 1$, while at all other integer fillings only single spin-flip excitations were observed. Other experiments using various experimental probes confirmed such findings and expanded on them \cite{14,15,16}. Measurements of the heat capacity in multiple quantum wells showed a sharp heat capacity peak at $T \sim 40$ mk near $\nu \sim 1$ which was interpreted as a possible transition between a liquid and lattice state of skyrmions \cite{13,14}. Our experiment addresses the limiting $T \to 0$ behavior near $\nu \sim 1$ by measuring the nuclear spin-lattice relaxation rate ($1/T_1$) using resistively detected NMR \cite{24,21,22,23,24,25}. Recently, Desrat \textit{et al.}
exploited this technique, and observed resistively detected NMR over a broad range of filling factors. In particular, they found an ‘anomalous’ dispersive-like line shape near $\nu \sim 1$, for which they speculated as originating from a possible coupling to a Skyrmie crystal.

The experiment was performed on a 40 nm wide modulation-doped GaAs/AlGaAs quantum well grown by molecular beam epitaxy. The electron density of the 2DEG was determined to be $n = 1.60(1) \times 10^{11}$ cm$^{-2}$, and the mobility $\mu \simeq 17 \times 10^6$ cm$^2$/V·s. Cooling of the electrons down to temperatures near the base temperature ($\sim 20$ mK) was achieved by thermally anchoring the sample leads on the refrigerator by means of various powder and RC filters with low cutoff frequencies. The temperature was determined with a ruthenium oxide thermometer. Figure 1 shows an example of magnetotransport measurement at $T \simeq 20$ mK.

A cartoon of our experiment is shown in Fig.1. An NMR coil is wrapped around the sample which resides in a strong ($\sim 8$T) perpendicular field $H_0$. A small radio-frequency (RF) field $|H_1 \cos(\omega t)| \sim \mu T$ matching the NMR frequency $f_{NMR} = \gamma H_0$ is radiated on the sample, where $\gamma = 7.29$ MHz/T for the $^{75}\text{As}$ nuclei. The resistance, $R_{xx}$, is monitored at constant $H_0$ and $T$ while the RF-field if slowly swept across the nuclear resonance at a rate of $\sim 0.15$ kHz/s. A typical resistively detected NMR signal for the $^{75}\text{As}$ nucleus is shown in the central inset of Fig.1 at filling factor $\nu = 0.86$. A small, but sizable resistance change $\delta R_{xx}$ is observed at resonance having typical signal strength $\delta R_{xx}/R_{xx} \sim 1\%$. The detection scheme in resistive NMR relies on the hyperfine interaction $\mathcal{A} \mathbf{I} \cdot \mathbf{S}$. In GaAs, the electronic Zeeman energy for the 2DEG can be written as $E_z = g^* \mu_B (H_0 + B_N)$, where $B_N = A < I_z > / g^* \mu_B$ is the Overhauser shift, $A$ the hyperfine constant and $< I_z >$ the nuclear spin polarization. Applying an RF-field at resonance modifies the thermal distribution of the nuclear spins in the applied field $H_0$ which depolarizes the nuclear spins and as a consequence modifies the Zeeman gap, $\Delta_z = g^* \mu_B (H_0 + B_N)$. For odd integer filling factors such as near $\nu \sim 1$, the electronic transport in the thermally activated regime is given by $R_{xx} \sim e^{-t/T_1}$ which makes possible the detection of the NMR by means of resistivity. A study and a discussion of the lineshape will be published elsewhere.$^{20}$

The inset of Fig.2 shows an example of spin-lattice relaxation time $T_1$ measurements. The resistance of the 2DEG is initially measured at constant field $H_0$ and temperature $T$ and monitored in time with the frequency set to be off-resonance (see the arrows in the central inset of Fig.1). At the time labeled ‘1’, the frequency is moved on-resonance (see Fig.1) and the resistance decreases as a consequence of the nuclei being depolarized, and eventually reaches a steady state. At ‘2’, the frequency is set back to off-resonance, and the resistance decays to its original state as the macroscopic nuclear magnetization $\mathcal{M}(t)$ relaxes in a time $T_1$ to its thermal equilibrium value, $\mathcal{M}_0$. The time dependence of $R_{xx}(t)$ is found to fit very well a single exponential of the form $R_{xx}(t) = \alpha + \beta e^{-t/T_1}$ (solid line in the inset of Fig.2). Here, $T_1$, is the characteristic relaxation
values. At temperatures 

time of the resistance and \( \alpha, \beta \) are coefficients which 
determine the on-resonance and off-resonance resistance 
values. At temperatures \( T \gtrsim 30 \) mK, the maximum 
change in the Zeeman gap between on-resonance and 
off-resonance is \( (\delta \Delta_z)^{max} = g' \mu_B B_N \) which is smaller 
than \( 2k_B T \) by at least a factor of 4. In addition, the \( T_1 \) 
measurements were performed in the small-power limit 
such that \( (\delta R_{xx})_{low-power} \lesssim 0.1 \cdot (\delta R_{xx})_{max} \), i.e. with 
partially depolarized nuclei only. Therefore, \( \delta \Delta_z \sim \delta B_N \ll 2k_B T \), and to first order the resistance scales as 
\( \delta R_{xx} \propto \frac{2 \mu_B B_N}{2k_B T} \). Noting that \( B_N \propto M \), we find 
\( T_{1l} \approx T_1 \) to a very good approximation.

The main panel of Fig. 2 shows the temperature 
dependence of the spin-lattice relaxation times \( T_1 \) for the 
\(^{75}\)As nuclei at filling factors \( \nu = 0.84 \) (diamonds), 0.86 
(filled circles) and 0.895 (empty circles). The temperature 
quoted is to a very good approximation the actual 

temperature, \( T_s \). This was determined by using the electronic resistance as an in situ thermometer. 
The x-axis error bars are remaining uncertainties in 

determining this temperature. Each \( T_1 \) datum was re-
produced over at least three independent measurements. 
The y-axis error bar provides a range for the scattering at 

each data point. No systematic dependence of \( T_1 \) on the 
RF-power was observed.

The relaxation time decreases significantly as \( T \to 0 \) 
for the three filling factors. The data at \( \nu = 0.895 \) shows 
a similar behaviour near \( T \approx 130 \) mK, at which point 
we could no longer detect the NMR signal. These data 
show that the nuclear spins relax much more efficiently 
as \( T \to 0 \) and that strong magnetic fluctuations exist 
in the quantum many-body ground state. Interestingly,

\[ T_1 \] also becomes shorter with \( \nu \) increasing. Barrett et al. 
showed that the nuclear spin relaxation was strongly 
suppressed for filling factor at, or very close to \( \nu = 1 \) and 
at \( T \sim 1 \) K. Our data are outside of this regime, 
\( |\nu - 1| \lesssim 0.1 \), and it is difficult to address this filling factor 
region with resistive NMR since the resistance vanishes 
in the quantum Hall state. Nevertheless, we expect \( T_1 \) 
to become much longer when the first Landau level is 
completely filled and the resulting ferromagnetic state 
suppresses the spin degree-of-freedom.

Inspecting the data of Fig. 2 one wonders as to the 
origin of this strong T-dependence of \( T_1 \). In fact, given that 
\( R_{xx} \) itself is strongly T-dependent and \( \nu \)-dependent the 
relationship seen in Fig. 2 may reflect a correlation with 
\( R_{xx} \). Towards this end we note that in the quantum Hall 
regime vanishing \( R_{xx} \) implies vanishing conductivity owing 
to the tensor inversion, \( \sigma_{xx} = \rho_{xx}/(\rho_{xx}^2 + \rho_{xy}^2) \), and a 
finite quantized diagonal resistivity, \( \rho_{xy} \). This insulating 
behavior in the quantum Hall regime is a consequence of 
the localization of the electrons (holes) by disorder with 
density \( \nu^* = \nu - K \text{ (} < 0 \text{ for holes)} \). Here, \( K \) is an integer 
which equals one at \( \nu = 1 \). Recent micro-wave measure-
ments performed on similar high-mobility samples show 
pinning resonances which suggest that a collective 
electron solid is formed in this regime.

Figure 3 shows the spin-lattice relaxation rate \( (1/T_1) \) 
of Fig. 2 plotted versus \( R_{xx} \). It shows a remarkable linear 
relationship linking an increase in relaxation rate with 
decreasing \( R_{xx} \) and hence with increasing localization. 
This strongly suggests that the nuclear spin relaxation is 
induced predominantly by those electrons (or holes) 
forming a solid, rather than the remaining conducting 
electronic states contributing to \( \sigma_{xx} \). It is important to 
realize that the data follow a trend opposite to the usual 
Korringa behavior observed in metals, \( (1/T_1) \propto N_F \propto \sigma_{xx} \propto \rho_{xx} \). 
\( N_F \) is the electronic density of states. If the 
Korringa relation were to hold, one would expect stronger 
relaxation at higher values of \( R_{xx} \), opposite to our data.

Efficient relaxation of the nuclear spins requires mag-
netic fluctuations in their environment. The data in Fig. 3 
requires an increase of such fluctuations as \( R_{xx} \) (and 
hence \( \sigma_{xx} \)) decreases. A spin-polarized two-dimensional 
Fermi gas is extremely inefficient in providing such fluctu-
ations. For instance, we measured the spin-lattice relax-
ation time \( T_1 \) in the high-field phase \( (B \gtrsim 30T) \) at small 
filling factors \( \nu \lesssim \frac{2}{3} \) and \( \nu \lesssim \frac{1}{3} \) where a Wigner crystal 
phase of electrons is expected to form. In contrast to 
near \( \nu \approx 1 \), \( T_1 \) in the Wigner crystal regime was found 
to be very long, ranging from \( \sim 550 \) to 1000 s at \( T \sim 50 \) 
mK and for all values of \( R_{xx} \). On the other hand, a spin-
wave Goldstone mode of a Skyrme crystal provides a 
very efficient mechanism for relaxing the nuclear spins. 
At magnetic fields \( H_0 \approx TT \) near \( \nu \approx 1 \), our sample has a 
Zeeman-to-Coulomb energy ratio \( \eta \approx 0.015 \) which favors 
Skyrmion formation and well below the \( \eta_c \approx 0.022 \) where 
they are expected to disappear. Such a Skyrme crys-
tal was calculated to enhance the nuclear spin relaxation by a factor $\sim 10^4$ over that of a 2D Fermi gas. This is consistent with the $\sim 10^4 - 10^6$ increase in relaxation that we observed near $\nu \sim 1$ as compared to the rate in the high-field electron solid phase. In addition, we have measured $T_1$ in the same sample around $\nu = 3$, and found that $T_1 \gtrsim 300 \text{ s}$ for $T \to 0$ and $R_{xx} \to 0$ in contrast to our result near $\nu \sim 1$. This is consistent with the result by Schmeller et al. [16] which showed no Skyrmion formation to occur at $\nu \sim 3$.

Extrapolating the rate in Fig.3 to the x-axis defines a resistance $R_m$ at which $T_1 \to \infty$. While there will remain other, weaker relaxation mechanisms, this extrapolated $R_m$ should provide a measure where nuclear relaxation by the strong low-temperature mechanism ceases. When translated to $T_m$ via our $R_{xx}$ versus $T$ calibration, the $T_m$’s define a temperature boundary for the low-temperature magnetic phase as shown in the inset of Fig.3 versus the partial filling factor, $|\nu - 1|$. The error bars correspond to errors in determining $R_m$ from extrapolating to $(1/T_1) \to 0$. The solid line in the inset is a guide-to-the-eye and the dotted line is an extrapolation to zero temperature. The shaded region is a proposed partial phase diagram spanned by the low-temperature magnetic phase (phase 1). The curvature of the data suggests the existence of a critical filling factor $\nu_m \sim 0.83$ defining a quantum phase transition between phases 1 and 2. A similar phase diagram has been deduced in the same sample around $\nu = 3$. While there will remain other, weaker relaxation mechanisms, this extrapolation to (1/$T_1$)→0. The solid line in the inset is a guide-to-the-eye and the dotted line is an extrapolation to zero temperature. The shaded region is a proposed partial phase diagram spanned by the low-temperature magnetic phase (phase 1). The curvature of the data suggests the existence of a critical filling factor $\nu_m \sim 0.83$ defining a quantum phase transition between phases 1 and 2. A similar phase diagram has been deduced in the vicinity of the $\nu = 3$ quantum Hall state, the nuclear spin relaxation increases strongly as the temperature is lowered and when $R_{xx} \to 0$. This strongly suggests that the localized states are responsible for the fast nuclear relaxation. We find a natural interpretation of our data in terms of a magnetic phase of localized skyrmions relaxing the nuclear spin via a Goldstone mode of the crystal and deduce a partial phase diagram in the $T - \nu$ plane.

The authors would like to acknowledge helpful discussions with Yong Chen, Lloyd Engel, Herb Fertig, Michael Hilke, René Côté, Mansour Shayegan and Kun Yang. One of the authors (G.G.) is grateful for the hospitality of the National High Magnetic Field Laboratory. Research funded by the NSF under grant # DMR-0084173 and # DMR-03-52738, and by the DOE under grant # DE-AIO2-04ER46133.

[1] T.H.R. Skyrme, Nucl. Phys. 31, 556 (1962), and references therein.
[2] S.L. Sondhi, A. Karihede, S.A. Kivelson and E.H. Rezayi, Phys. Rev. B 47, 16419 (1993).
[3] L. Brey, H.A. Fertig, R. Côté and A.H. MacDonald, Phys. Rev. Lett. 75, 2562 (1995).
[4] I. Klebanov, Nucl. Phys. B 262, 133 (1985).
[5] V. Bayot, E. Grivei, S. Melinte, M.B. Santos and M. Shayegan, Phys. Rev. Lett. 75, 4598 (1995).
[6] V. Bayot, E. Grivei, J.-M. Beuken, S. Melinte and M. Shayegan, Phys. Rev. Lett. 79, 1718 (1997).
[7] R. Côté, A.H. MacDonald, Luis Brey, H.A. Fertig, S.M. Girvin, and H.T.C Stoof, Phys. Rev. Lett. 78, 4825 (1997).
[8] H.A. Fertig, L. Brey, R. Côté, and A.H. MacDonald, Phys. Rev. B 50, 11018 (1994).
[9] H.A. Fertig, L. Brey, R. Côté, A.H. MacDonald, A. Karihede, and S.L. Sondhi, Phys. Rev. B 55, 10671 (1997).
[10] C. Timm, S.M. Girvin, and H.A. Fertig, Phys. Rev. B 58, 16334 (1998).
[11] A.G. Green, Phys. Rev. B 61, R16299 (2000).
[12] J. Sinova, S.M. Girvin, T. Jungwirth, and K. Moon, Phys. Rev. B 61, 2749 (2000).
[13] B. Paredes and J.J. Palacios, Phys. Rev. B 60, 15570 (1997); J.P. Rodriguez, Europhys. Lett. 42, 197 (1998).
[14] S.E. Barrett, G. Dabbagh, L.N. Pfeiffer, K.W. West and R. Tycko, Phys. Rev. Lett. 74, 5112 (1995).
[15] R. Tycko, S.E. Barrett, G. Dabbagh, L.N. Pfeiffer and K.W. West, Science 268, 1460 (1995).
[16] A. Schmeller, J.P. Eisenstein, L.N. Pfeiffer and K.W. West, Phys. Rev. Lett. 75, 4290 (1995).
[17] S. Melinte, N. Freytag, M. Horvatić, C. Berthier, L.P. Lévy, V. Bayot and M. Shayegan, Phys. Rev. B 64, 085327 (2001).
[18] P. Haldelwal, A.E. Dementyev, K.N. Kuzma, S.E. Barrett, L.N. Pfeiffer and K.W. West, Phys. Rev. Lett. 86, 5353 (2001).
[19] J.G. Groshaus, V. Umansky, H. Shtrikman, Y. Levinson, and I. Bar-Joseph, Phys. Rev. Lett. 93, 096802 (2004).
[20] M. Dobers, K. v. Klitzing, J. Schneider, G. Weinmann.
and K. Ploog, Phys. Rev. Lett. 61, 1650 (1988).
[21] S. Kronmuller, W. Dietsche, K. v. Klitzing, G. Denninger, W. Wegscheider and M. Bichler, Phys. Rev. Lett. 82, 4070 (1999).
[22] J.H. Smet, R.A. Deutschmann, F. Ertl, W. Wegscheider, G. Abstreiter and K. v. Klitzing, Nature (London) 415, 281 (2002).
[23] W. Desrat, D.K. Maude, M. Potemski, J.C. Portal, Z.R. Wasilewski and G. Hill, Phys. Rev. Lett. 88, 256807 (2002).
[24] K. Hashimoto, K. Muraki, T. Saku, and Y. Hirayama, Phys. Rev. Lett. 88, 176601 (2002).
[25] O. Stern, N. Freytag, A. Fay, W. Dietsche, J. H. Smet, K. von Klitzing, D. Schuh, and W. Wegscheider, Phys. Rev. Lett. 70, 075318 (2004).
[26] G. Gervais, H.L. Stormer, D.C. Tsui, P.L. Kuhns, W.H. Moulton, A.P. Reyes, L.N. Pfeiffer, and K.W. West, to be published.
[27] Yong Chen, R.M. Lewis, L.W. Engel, D.C. Tsui, L.N. Pfeiffer, and K.W. West, Phys. Rev. Lett. 91, 016801 (2003).