Evidence for Skyrmion Crystallization from NMR Relaxation Experiments

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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 $T \rightarrow 0$ and when a well developed quantum Hall state with $R_{xx} \simeq 0$ 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.

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In his seminal work on nuclear matter more than 40 years ago Skyrme showed that baryons emerge mathematically as a static solution of a meson field described by the so-called Skyrme Lagrangian [1]. His work provided the foundation for the quantum theory of solitons, and more recently found an interesting and *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 levels is almost completely occupied, the elementary excitations of the system become large topologically stable spin textures known as Skyrmions [2]. It was further proposed that at $T = 0$ Skyrmions would localize on a square lattice [3]. This ground state represents a new type of magnetic ordering which possesses long-range orientation and positional order, and is the solid-state analogue of the Skyrmion crystal state which is used to describe dense nuclear matter [4]. In this Letter, 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 $T \rightarrow 0$ and $R_{xx} \rightarrow 0$ where localization of electronic states occurs. This is consistent with previous measurements of the heat capacity in GaAs/AlGaAs at very low temperatures [5,6], previous NMR work [7], and with the predictions of a Skyrmion crystal [8].

Several theoretical publications [2,3,8–13] have pointed toward the existence of Skyrmions in a two-dimensional electron gas (2DEG). At filling factor $\nu = 1$, where ν is defined by the ratio of the electronic density n to the magnetic flux density, $\nu = \frac{n}{B/\Phi} = \frac{nh}{eB}$, the quantized Hall state is ferromagnetic. For sufficiently small Zeeman-to-Coulomb energy ratio $\eta = E_z/E_c = \frac{g^* \mu_B B}{e^2/\epsilon l_B}$, where g^* is the electronic g factor and $l_B = \sqrt{\hbar/eB}$ is the magnetic length, Sondhi *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 [2]. These Skyrmions are topologically stable, charged $\pm e$, and gapped excitations which are the result of an energy trade off 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 [3,11,12] it is theoretically debated [14] 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 *et al.* showed strong evidence for a finite density of Skyrmions at temperature $T \sim 1$ K [15,16]. Subsequently, Schmeller *et al.* [17] 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 [18–20]. 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 [5,6]. Our experiment addresses the limiting $T \rightarrow 0$ behavior near $\nu \sim 1$ by measuring the nuclear spin-lattice relaxation rate $(1/T_1)$ using resistively detected NMR [7,21–25]. Recently, Desrat *et al.* [7] exploited this technique, and observed an "anomalous" dispersivelike line shape near $\nu \sim 1$ for which they speculated as originating from a possible coupling to a Skyrme 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/Vs. Cooling of the electrons down to temperatures near the base temperature

(~ 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 (~ 8 T) perpendicular field H_0. A small radio-frequency (rf) field $|H_1 \cos(\omega t)| \sim \mu T$ matching the NMR frequency $f_{\mathrm{NMR}} = \gamma H_0$ is radiated on the sample, where $\gamma = 7.29$ MHz/T for the ^{75}As nuclei. The resistance, R_{xx}, is monitored at constant H_0 and T while the rf field is slowly swept across the nuclear resonance at a rate of $\sim$0.15 kHz/s. A typical resistively detected NMR signal for the ^{75}As nucleus is shown in the central inset of Fig. 1 at filling factor $\nu = 0.86$, with a typical signal strength $\delta R_{xx}/R_{xx} \sim 1\%$. A discussion of the coupling between R_{xx} and the nuclear resonance can be found in Ref. [7]. Near $\nu \sim 1$, we have also observed under some restrictive conditions (high rf levels) a dispersivelike, non-Lorentzian line shape similar to those observed by Desrat *et al.* [7]. A study and a discussion of the line shape will be published elsewhere [26].

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 $R_{xx}(t) = \alpha + \beta e^{-t/T_{1\prime}}$ (solid line in the inset of Fig. 2) and with a single relaxation time, $T_{1\prime}$. Here, $T_{1\prime}$ is the characteristic relaxation time of the resistance and α, β are coefficients which determine the resistance amplitudes. 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 $(\delta R_{xx})_{\text{low power}} \lesssim 0.1(\delta R_{xx})_{\max}$, 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 $\delta R_{xx} \propto \frac{g^* \mu_B \delta B_N}{2k_B T}$. Noting that $B_N \propto \mathcal{M}$, we find $T_{1\prime} \simeq T_1$ to a very good approximation. At filling factors away from $\nu = 1$, we found no significant deviation from a single exponential for the resistance recovery, suggesting that our approximation $T_{1\prime} \simeq T_1$ remains valid.

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 electronic temperature, T_e. 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 reproduced over at least three

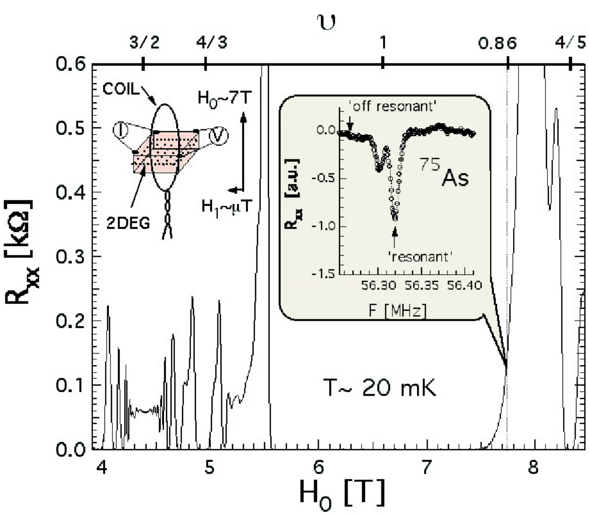

FIG. 1 (color online). Magnetotransport R_{xx} for the two-dimensional electron system. The bottom x axis shows the applied magnetic field and the upper x axis the corresponding filling factors. A schematic of the NMR experiment is shown in the upper left corner. The inset at the center of the graph shows an example of resistively detected NMR resonance for the ^{75}As nucleus and at filling factor $\nu = 0.86$.

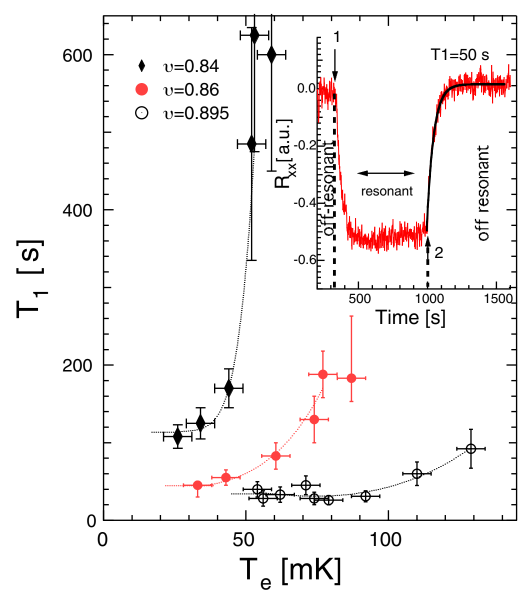

FIG. 2 (color online). Nuclear spin-lattice relaxation time T_1 for ^{75}As at various electronic temperatures T_e and filling factors, ν. The thin dotted lines are guides to the eye. The inset shows a typical experiment from which T_1 is determined (see text). The solid line is a fit to a single exponential.

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 behavior near $T \sim 130$ 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. Similar reductions in T_1 in the same filling factor range have been observed previously in Ref. [23,24]. Interestingly, T_1 also becomes shorter with ν 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 [15,16]. 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 ν 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, ρ_{xy}. This insulating behavior in the quantum Hall regime is a consequence of the localization of the electrons (holes) by disorder with density $\nu^* = \nu - K$ (< 0 for holes). Here, K is an integer which equals one at $\nu = 1$. Recent microwave measurements performed on similar high-mobility samples show pinning resonances which suggest that a collective electron solid is formed in this regime [27].

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 σ_{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.

The data in Fig. 3 requires an increase in magnetic fluctuations as R_{xx} (and hence σ_{xx}) decreases. A spin-polarized two-dimensional Fermi gas is extremely inefficient in providing such fluctuations. For instance, we measured the spin-lattice relaxation time T_1 in the high-field phase ($B \gtrsim 30T$) at small filling factors $\nu < \frac{2}{9}$ and $\nu < \frac{1}{5}$ where a Wigner crystal phase of electrons is expected to

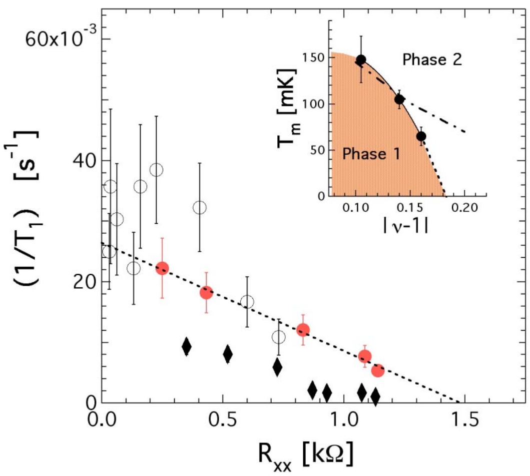

FIG. 3 (color online). Nuclear spin-lattice relaxation rate $(1/T_1)$ for ^{75}As versus R_{xx} at the same filling factors as in Fig. 2. The straight line is a linear fit to the $\nu = 0.86$ data. T_m (defined in the text) is shown in the inset (filled circles) versus the partial filling factor, $|\nu - 1|$. The solid line is a guide to the eye, and the dotted line an extrapolation to $T = 0$. The dash-dotted line is a theoretical estimate for the classical transition temperature of a Skyrme crystal [8] (see text).

form [26]. In contrast to near $\nu \sim 1$, T_1 in the Wigner crystal regime was found to be very long, ranging from $\sim$350 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 [8] provides a very efficient mechanism for relaxing the nuclear spins. At magnetic fields $H_0 \sim 7T$ near $\nu \sim 1$, our sample has a Zeeman-to-Coulomb energy ratio $\eta \simeq 0.015$, which favors Skyrmion formation, and well below the $\eta_c \simeq 0.022$ where they are expected to disappear [9]. Such a Skyrmion crystal was calculated to enhance the nuclear spin relaxation by a factor $\sim 10^3$ over that of a 2D Fermi gas. This is consistent with the $\sim 10^1$–10^2 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 $T_1 \gtrsim 300$ 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.* [17] 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 phase boundary, 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 $T = 0$. 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 in the range $\nu_m \sim 0.80$–0.83 defining a quantum phase transition between phases 1 and 2. A similar phase diagram has been deduced in the heat capacity work of Bayot *et al.* [5,6]. While in their work the overall values of T_m are smaller, we expect the transition to be sensitive to disorder so the discrepancy may simply reflect the overall lower disorder in our sample.

An estimate for the classical melting temperature of a Skyrme crystal can be made from the relation $T_{\rm crystal} = (b^2\mu/4\pi)$. The shear modulus μ of the Skyrme crystal can be calculated from a microscopic theory, which together with the lattice constant b yield $T_{\rm crystal} \sim 0.363$ K at $|\nu - 1| \sim 0.85$ [8]. Similar calculations for the melting of an electron crystal usually overestimate the melting transition by a factor of 2, and quantum positional fluctuations are expected to further suppress it. Using the value for $T_{\rm crystal}$ given in Ref. [8], we plotted its partial filling factor dependence in the inset of Fig. 3 with a dash-dotted line. Its magnitude was rescaled by a factor of 3.6 so as to match the overall magnitude of our data. While the trend of the data is not completely accounted for, the model, indeed, predicts T_m decreasing with increasing $|\nu - 1|$. Within this scenario, phase 1 would correspond to a square lattice phase of Skyrmions with long-range positional and orientation order, while phase 2 would correspond possibly to a melted Skyrmion phase with quasi-long-range magnetic order only.

In conclusion, we presented a detailed study of the nuclear spin-lattice relaxation rate near $\nu = 1$ in the $T \rightarrow 0$ limit of an extremely high-quality GaAs/AlGaAs sample. In the vicinity of the $\nu = 1$ quantum Hall state, the nuclear spin relaxation increases strongly as the temperature is lowered and when $R_{xx} \rightarrow 0$. 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.

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[1] T. H. R. Skyrme, Nucl. Phys. **31**, 556 (1962), and references therein.
[2] S.L. Sondhi, A. Karlhede, S.A. Kivelson, and E.H. Rezayi, Phys. Rev. B **47**, 16 419 (1993).
[3] L. Brey, H.A. Fertig, R. Côté, and A.H. MacDonald, Phys. Rev. Lett. **75**, 2562 (1995).
[4] I. Klebanov, Nucl. Phys. **B262**, 133 (1985).
[5] V. Bayot, E. Grivei, S. Melinte, M.B. Santos, and M. Shayegan, Phys. Rev. Lett. **76**, 4584 (1996).
[6] V. Bayot, E. Grivei, J.-M. Beuken, S. Melinte, and M. Shayegan, Phys. Rev. Lett. **79**, 1718 (1997).
[7] W. Desrat, D.K. Maude, M. Potemski, J.C. Portal, Z.R. Wasilewski, and G. Hill, Phys. Rev. Lett. **88**, 256807 (2002).
[8] R. Côté, A.H. MacDonald, Luis Brey, H.A. Fertig, S.M. Girvin, and H.T.C Stoof, Phys. Rev. Lett. **78**, 4825 (1997).
[9] H.A. Fertig, L. Brey, R. Côté, and A.H. MacDonald, Phys. Rev. B **50**, 11 018 (1994).
[10] H.A. Fertig, L. Brey, R. Côté, A.H. MacDonald, A. Karlhede, and S.L. Sondhi, Phys. Rev. B **55**, 10 671 (1997).
[11] C. Timm, S.M. Girvin, and H.A. Fertig, Phys. Rev. B **58**, 10 634 (1998).
[12] A.G. Green, Phys. Rev. B **61**, R16 299 (2000).
[13] J. Sinova, S.M. Girvin, T. Jungwirth, and K. Moon, Phys. Rev. B **61**, 2749 (2000).
[14] B. Paredes and J.J. Palacios, Phys. Rev. B **60**, 15 570 (1999); J.P. Rodriguez, Europhys. Lett. **42**, 197 (1998).
[15] S.E. Barrett, G. Dabbagh, L.N. Pfeiffer, K.W. West, and R. Tycko, Phys. Rev. Lett. **74**, 5112 (1995).
[16] R. Tycko, S.E. Barrett, G. Dabbagh, L.N. Pfeiffer, and K.W. West, Science **268**, 1460 (1995).
[17] A. Schmeller, J.P. Eisenstein, L.N. Pfeiffer, and K.W. West, Phys. Rev. Lett.**75**, 4290 (1995).
[18] S. Melinte, N. Freytag, M. Horvatić, C. Berthier, L.P. Lévy, V. Bayot, and M. Shayegan, Phys. Rev. B **64**, 085327 (2001).
[19] P. Khandelwal, A.E. Dementyev, K.N. Kuzma, S.E. Barrett, L.N. Pfeiffer, and K.W. West, Phys. Rev. Lett. **86**, 5353 (2001).
[20] J.G. Groshaus, V. Umansky, H. Shtrikman, Y. Levinson, and I. Bar-Joseph, Phys. Rev. Lett. **93**, 096802 (2004).
[21] M. Dobers, K.v. Klitzing, J. Schneider, G. Weinmann, and K. Ploog, Phys. Rev. Lett. **61**, 1650 (1988).
[22] S. Kronmuller, W. Dietsche, K.v. Klitzing, G. Denninger, W. Wegscheider and M. Bichler, Phys. Rev. Lett. **82**, 4070 (1999).
[23] J.H. Smet, R.A. Deutschmann, F. Ertl, W. Wegscheider, G. Abstreiter, and K.v. Klitzing, Nature (London) **415**, 281 (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.v. Klitzing, D. Schuh, and W. Wegscheider, Phys. Rev. B **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).