Heat Capacity Evidence for the Suppression of Skyrmions at Large Zeeman Energy

S. Melinte\textsuperscript{1}, E. Grivei\textsuperscript{1}, V. Bayot\textsuperscript{1} and M. Shayegan\textsuperscript{2}

\textsuperscript{1}Unité PCPM, Université Catholique de Louvain, 1348 Louvain-la-Neuve, Belgium
\textsuperscript{2}Department of Electrical Engineering, Princeton University, Princeton N.J. 08544

(March 24, 2022)

Abstract

Measurements on a multilayer two-dimensional electron system (2DES) near Landau level filling $\nu=1$ reveal the disappearance of the nuclear spin contribution to the heat capacity as the ratio $\tilde{g}$ between the Zeeman and Coulomb energies exceeds a critical value $\tilde{g}_c \approx 0.04$. This disappearance suggests the vanishing of the Skyrmion-mediated coupling between the lattice and the nuclear spins as the spin excitations of the 2DES make a transition from Skyrmions to single spin-flips above $\tilde{g}_c$. Our experimental $\tilde{g}_c$ is smaller than the calculated $\tilde{g}_c=0.054$ for an ideal 2DES; we discuss possible origins of this discrepancy.

PACS numbers: 73.20.Dx, 73.40.Hm, 65.40.-f
The ground state and spin excitations of a two-dimensional electron system (2DES) near Landau level (LL) filling $\nu=1$ have attracted much recent interest [1–9]. At this filling, the Coulomb exchange energy plays a dominant role, leading to a substantially larger quantum Hall effect (QHE) excitation gap than the expected single-particle Zeeman splitting [1]. Moreover, the lowest energy charged excitations of the 2DES at $\nu=1$ are expected to be spin textures known as Skyrmions [2], provided that the Zeeman energy is small. Pioneering nuclear magnetic resonance (NMR) experiments revealed a pronounced reduction of the 2DES spin polarization and a strong enhancement of the nuclear spin-lattice relaxation rate near $\nu=1$ [3]. These observations have been attributed to the presence of Skyrmions in the electronic ground state [4], and to Skyrmion-induced low-energy spin-flip excitations [5], respectively. Magnetotransport [6,7] and magnetooptical [8] data have provided further evidence for the existence of finite-size Skyrmions.

Low temperature ($T$) heat capacity ($C$) measurements on a GaAs/AlGaAs multiple-quantum-well (MQW) heterostructure revealed that $C$ near $\nu=1$ is dominated by the Schottky nuclear heat capacity [10] of Ga and As atoms in the quantum wells (QWs). This observation implies a strong enhancement of the nuclear spin-lattice relaxation rate near $\nu=1$, consistent with NMR experiments [3], and suggests that heat capacity is a sensitive probe of the presence or the absence of Skyrmions in 2DESs.

Hitherto, most of the experimental studies at $\nu=1$ concentrated on the range of small ratio $\tilde{g}=|g^*|\mu_B B/(e^2/\epsilon l_B)$ between the Zeeman and Coulomb energies [3,7,8,10], where $|g^*|\mu_B=0.3$ K/T, $\epsilon\approx 13\epsilon_0$, $l_B=(h/eB_\perp)^{1/2}$ is the magnetic length, and $B$ and $B_\perp$ are the total and perpendicular components of the magnetic field, respectively. One important theoretical prediction is that a transition from Skyrmions to single spin-flip excitations is expected above a critical $\tilde{g}_c$ [2,9]. Up to now, the experiments performed at large $\tilde{g}$ probed the QHE activation energy at $\nu=1$ ($\Delta$) which exhibits only a smooth and gradual evolution with $\tilde{g}$ [2]. This fact prohibits an accurate determination of $\tilde{g}_c$ and restricts the investigations to the case of filling exactly equal to unity. In this paper, we report evidence for $\tilde{g}_c$ in the range $0.7 \lesssim \nu \lesssim 1.3$ in a low-density, modulation-doped GaAs/AlGaAs MQW heterostructure.
The heat capacity near $\nu=1$ and $\Delta$ at $\nu=1$ were measured as the Zeeman energy was tuned by tilting the sample in the magnetic field. The data show that the nuclear contribution to the heat capacity decreases in a narrow $\tilde{g}$-range and is suppressed for $\tilde{g}>0.04$. This result provides evidence for a rather abrupt transition from Skyrmions to single spin-flip excitations at $\tilde{g}_c \approx 0.04$. This $\tilde{g}_c$ is somewhat smaller than $\tilde{g}_c=0.054$ expected from theoretical calculations for an ideal 2D system [2,9]. The discrepancy likely comes from corrections to $\tilde{g}_c$ because of finite layer-thickness, LL mixing, and disorder in a real sample. We also discuss the possibility of a LL crossing at high tilt angles ($\theta$) which might be responsible for the suppression of Skyrmions in our sample for $\tilde{g} \geq 0.04$. Finally, the heat capacity data at intermediate $\theta$ ($0.02 \leq \tilde{g} \leq 0.03$) exhibits some anomalous features which we presently do not understand.

The heterostructure used in this study consists of one hundred 300 Å-thick GaAs QWs, separated by 2500 Å-thick Al$_{0.1}$Ga$_{0.9}$As barriers which are δ-doped with donors (Si) near their centers. Heat capacity experiments were carried on a 7×10 mm$^2$ piece of the wafer thinned to 160 µm. The thermometer and heater are carbon paint resistors, deposited on the substrate side of the sample and electrically connected with graphite fiber bundles which also serve as thermal link to the heat sink. In the restricted $T$ and $\nu$-range investigated here, we used a corrected relaxation method [10,11] for $C$-experiments: $C = \kappa \tau_{ext}$, where $\kappa$ and $\tau_{ext}$ are the measured thermal conductance to the heat sink and the external time constant, respectively. It is important to note that we measure the heat capacity of those nuclei with a nuclear spin-lattice relaxation time $T_1 < \tau_{ext}$. Typical $\tau_{ext}$ in our experiments was $\sim 100$ to 1000 s. We also performed electrical measurements on a 2×2 mm$^2$ piece from the same wafer. Both measurements were performed in a dilution refrigerator and the samples were tilted in situ so that an angle $0^\circ \leq \theta \leq 77^\circ$ forms between $B$ and the normal to the sample plane.

Figure 1 shows the longitudinal resistance ($R_{xx}$) as a function of $B$ at $\theta=0^\circ$. The density determined from the position of $R_{xx}$ minima is $8.5 \times 10^{10}$ cm$^{-2}$ per layer and the estimated mobility is $\approx 7 \times 10^5$ cm$^2$/Vs. The very high quality of this heterostructure is evidenced by
the presence of strong fractional QHE states at ν = 2/5 and 3/5. The value of the excitation gap (Δ) was determined from the T-dependence of \( R_{xx} \) at ν = 1 in the thermally activated regime where \( R_{xx} \propto \exp(-\Delta/2T) \). At \( \theta = 0^\circ \), Δ = 20 K, comparable to the measured Δ in high-quality conventional single-layer 2DESs \[6\]. The evolution of Δ with \( \theta \) is presented in Fig. 2 by plotting Δ vs Zeeman energy, both expressed in units of \( e^2/\epsilon lB \). The inset to Fig. 2 shows the calculated Hartree-Fock (HF) energy gap for the creation of a widely separated Skyrmion/Antiskyrmion pair as a function of \( \tilde{g} \) for our sample \[12\]. This calculation takes into account the finite thickness of the electron layers; the relevant parameter is \( w/l_B = 0.52 \) where \( w = 71 \) Å is the rms width of the self-consistently calculated subband wavefunction in each QW.

Similar to previous results for single QWs \[6\], there is an overall qualitative agreement between the measured and calculated gaps in Fig. 2 \[13\]. In particular, assuming that the slope \( K = \partial \Delta / \partial (|g^*|\mu_B B) \) gives the number of flipped electron spins within a single, charged excitation at ν = 1, both theory and experiment give \( K \cong 9 \) for \( \tilde{g} \approx 0.012 \) and \( K \cong 1 \) in the limit of large \( \tilde{g} \) \[15\]. However, given the experimental uncertainty in the measured gaps, the absence of quantitative agreement with theoretical predictions, and the fact that Δ is expected to slowly approach the single spin-flip dependence (\( K = 1 \)), prohibit an accurate determination of \( \tilde{g}_c \) based on transport measurements.

We now present heat capacity experiments, which reveal a dramatic and rather abrupt dependence on \( \tilde{g} \) and provide evidence for the disappearance of Skyrmions. In the investigated range of \( T \) and \( B \), the results can be understood by using the Schottky nuclear heat capacity model developed in Ref. \[10\]. The model assumes a strong coupling of the nuclear spin system in the QWs to the lattice near ν = 1 provided by low-energy spin excitations \[3,5\]. In the present sample, the Schottky nuclear heat capacity of Ga and As atoms in the QWs is estimated at: \( C_{QW} = 3.3 \times 10^{-11} B^2 T^{-2} \) (J/K) \[10\]. Therefore, the ratio (\( \xi \)) between the measured \( C \) and calculated \( C_{QW} \) provides the fraction of QWs nuclei which strongly couple to the lattice, and hence signals the presence of low-energy spin excitations in the 2DES attributed to Skyrmions \[2,5\].
Figures 3 and 4 capture the evolution of the heat capacity, represented by the parameter $\xi \equiv C/C_{QW}$, with tilt angle, at $T=60$ mK. At $\theta=0^\circ$ (Fig. 3), the data are qualitatively similar to those reported for a 100-period GaAs/Al$_{0.3}$Ga$_{0.7}$As heterostructure with a density of $1.4 \times 10^{11}$ cm$^{-2}$ per layer [4]. The decreasing density of Skyrmions as $\nu \to 1$ is responsible for the minimum observed in $\xi$ at $\nu \approx 1$ [2]. The non-vanishing $\xi$ at $\nu=1$ could arise from density inhomogeneities in such a large, thinned MQW sample. Far away from $\nu=1$ ($\nu \lesssim 0.7$ and $\nu \gtrsim 1.4$), $\xi$ is essentially zero as the 2DES is at fillings where Skyrmions are no longer relevant [2,4]. We note that $\xi$ at maxima reaches supraunitary values: $\xi \approx 1.3$. Besides the experimental accuracy ($\pm 15\%$) and uncertainty in well-width ($\pm 10\%$), the barriers' nuclei may enhance the measured $C$ due to the spread of the electron wave function from the QWs into the Al$_{0.1}$Ga$_{0.9}$As barriers which are only about 0.1 eV high.

As seen in Fig. 3, $\xi$ vs $\nu$ at $\theta=46^\circ$, is nearly identical to the $\theta=0^\circ$ data. On the other hand, at $\theta=71^\circ$, the data show a significant asymmetry with respect to the $\nu=1$ position and a broadening of the $\nu>1$ peak. For $\nu < 1$, $\xi$ at $\nu \approx 0.9$ is reduced by a factor of 2 when compared to the $\theta=0^\circ$ value and vanishes for $\nu \lesssim 0.8$. Most remarkable, however, is that the magnitude of $\xi$ at the $\nu>1$ peak is comparable to the $\theta=0^\circ$ data, implying a still strong coupling of the nuclei to the lattice. This is a particularly noteworthy observation as it highlights that the heat capacity is a very sensitive probe of the low-energy spin excitations, and therefore Skyrmions, in a regime where the transport data and calculations both reveal a small Skyrmion size ($K<3$) and a very weak dependence of $\Delta$ on $\tilde{g}$ (see Fig. 2 and its inset near $\tilde{g} \approx 0.035$ ($\theta=71^\circ$)).

When $\theta$ is further increased above $71^\circ$ only by few degrees (Fig. 4), the nuclear heat capacity decreases dramatically for all $\nu$. For $\theta > 74^\circ$, the nuclear heat capacity is no longer measurable up to the highest investigated tilt-angle ($77^\circ$). To bring into focus the evolution of the coupling between the nuclear spin system and the lattice with $\theta$ and $\tilde{g}$, we plot $\xi$ at $\nu > 1$ and $\nu < 1$ maxima vs $\tilde{g}$ (Fig. 5). The coupling due to low-energy spin excitations is progressively suppressed for $\tilde{g} \gtrsim 0.035$ and vanishes in the range $0.037 \lesssim \tilde{g} \lesssim 0.043$. We believe that this behavior provides evidence for the transition from Skyrmions to single spin-flip
excitations at \( \tilde{g}_c \approx 0.04 \) in our sample. This \( \tilde{g}_c \) is smaller than the theoretical \( \tilde{g}_c = 0.054 \) calculated for the Skyrmion to single spin-flip transition for an ideal 2DES \([2,4]\). Several factors, however, are expected to reduce \( \tilde{g}_c \) for a real 2DES. This includes the finite thickness of the electron layer \([9]\), LL mixing \([13]\), LL crossing \([17]\), and disorder. Indeed, calculations by Cooper \([4,12]\) (also see inset to Fig. 2) reveal that taking into account the finite \( z \)-extent of the 2DES alone leads to \( \tilde{g}_c = 0.047 \), closer to our experimental value. It is worth emphasizing, however, that all these calculations are performed at \( \nu = 1 \) while the heat capacity data of Figs. 4 and 5 provide values for \( \tilde{g}_c \) in the full \( \nu \)-range where Skyrmions are the expected ground state of the 2DES. In particular, we observe that \( \tilde{g}_c \) depends on filling factor and increases from \( \tilde{g}_c \approx 0.037 \) at \( \nu = 1.2 \) to \( \tilde{g}_c \approx 0.043 \) at \( \nu = 0.9 \).

Finally, we wish to report an anomalous and unexpected behavior for the measured heat capacity at intermediate tilt-angles \( (50^\circ \lesssim \theta \lesssim 66^\circ) \) which shows evidence for the complex dependence of Skyrmions on \( \theta \) and \( \tilde{g} \). As depicted in Fig. 6, we observe a reduction of \( \xi \) in a narrow \( \nu \)-range compared to the \( \theta = 0^\circ \) data. The vertical arrows in Fig. 6 point to the total magnetic field at which \( \xi \) is most significantly reduced. The anomaly moves to higher \( \nu \) with small increases in \( \theta \) (from \( \nu = 0.81 \) at \( \theta = 50^\circ \) to \( \nu \approx 1 \) at \( \theta = 66^\circ \)) and shows an increasing intensity when compared to the \( \theta = 0^\circ \) data-envelope, e.g., at \( \theta = 50^\circ \), \( \xi \) at \( \nu = 0.81 \) is reduced by a factor of \( \approx 3 \), while at \( \theta = 66^\circ \), \( \xi \) at \( \nu \approx 1 \) is essentially zero. The observed reduction of \( \xi \) shows that low-energy spin excitations are strongly affected or even suppressed, which would signal the disappearance of Skyrmions for limited \( \nu \) or \( B \)-ranges dependent on tilt-angle. Even though the exact behavior of the anomaly might be specific to our heterostructure, it reveals the subtle influence of \( \theta \) on spin excitations of 2DESs whose description will require further theoretical and experimental work. However, we note that the anomaly strongly affects the heat capacity at \( \nu = 1 \) in the range \( 61^\circ \lesssim \theta \lesssim 66^\circ \) which corresponds to the range of \( \theta \) where the activation energy at \( \nu = 1 \) exhibits the strongest departure from the HF calculations \([15]\). The suppression of Skyrmions at \( \nu = 1 \) in this range of \( \theta \) would certainly affect \( \Delta \), and possibly increase it up to the single spin-flip value (dashed line in Fig. 2). This might explain the somewhat anomalous behavior of \( \Delta \) vs \( \tilde{g} \) in the range.
0.02 \lesssim \tilde{g} \lesssim 0.03.

In conclusion, the heat capacity experiments reveal the subtle and critical influence of tilted magnetic fields on the ground and excited states of 2DESs near $\nu=1$ in GaAs/AlGaAs heterostructures. The data indicate the disappearance of low-energy spin excitations in the 2DES, which in turn provides evidence for the suppression of Skyrmions above a critical Zeeman energy ($\tilde{g}_c \approx 0.04$ in our sample).

The authors are much indebted to N.R. Cooper, T. Jungwirth and S.P. Shukla for numerical calculations. This work has been supported by NATO grant CRG 950328 and the NSF MRSEC grant DMR-9400362. V.B. acknowledge financial support by the Belgian National Fund for Scientific Research. The work performed in Louvain-la-Neuve was carried out under financial support of the programme "PAI" sponsored by the "Communauté Française de Belgique".

**Note Added.** - Since the submission of this Letter we performed NMR experiments on the same heterostructure [18]. The measurements indicate that Skyrmions are suppressed at $\nu=1$ ($70^\circ \lesssim \theta \lesssim 73^\circ$) above $\tilde{g}_c \approx 0.038$ which is totally consistent with the present heat capacity data. Moreover, the NMR results suggest that the LL crossing occurs at significantly larger tilt angles ($\theta > 80^\circ$) and hence does not affect the observed $\tilde{g}_c$. 
REFERENCES

[1] R.J. Nicholas et al., Phys. Rev. B 37, 1294 (1988); A. Usher et al., Phys. Rev. B 41, 1129 (1990).

[2] S.L. Sondhi, A. Karlhede, S.A. Kivelson and E.H. Rezayi, Phys. Rev. B 47, 16419 (1993); E.H. Rezayi, Phys. Rev. B 43, 5944 (1991); C. Kallin and B.I. Halperin, Phys. Rev. B 30, 5655 (1984); D.-H. Lee and C.L. Kane, Phys. Rev. Lett. 64, 1313 (1990); H.A. Fertig et al., Phys. Rev. B 50, 11018 (1994).

[3] S.E. Barrett et al., Phys. Rev. Lett. 74, 5112 (1995); R. Tycko et al., Science 268, 1460 (1995).

[4] L. Brey et al., Phys. Rev. Lett. 75, 2562 (1995).

[5] R. Côté et al. Phys. Rev. Lett. 78, 4825 (1997).

[6] A. Schmeller et al., Phys. Rev. Lett. 75, 4290 (1995).

[7] D.K. Maude et al., Phys. Rev. Lett. 77, 4604 (1996).

[8] E.H. Aifer et al., Phys. Rev. Lett. 76, 680 (1996).

[9] N.R. Cooper, Phys. Rev. B. 55, R1934 (1997); H.A. Fertig et al., Phys. Rev. B 55, 10671 (1997).

[10] V. Bayot et al., Phys. Rev. Lett. 76, 4584 (1996); V. Bayot et al., Phys. Rev. Lett. 79, 1718 (1997).

[11] E. Grivei et al. (unpublished).

[12] N.R. Cooper (private communication).

[13] As in Ref. [3], our measured Δ is only 25% to 40% of the theoretical value for an ideal 2DES.

[14] Compared to the higher density heterostructure [10], ξ shows narrower peaks in ν and
the Schottky $T$-dependence ($C \propto T^{-2}$) is followed down to $T=28$ mK. The previously reported sharp peak in $C$ vs $T$ \cite{10} is not observed which might signal that it occurs at lower temperatures in this lower density sample.

\cite{15} Note in Fig. 2 that near $\bar{g}=0.025$ ($61^\circ < \theta < 66^\circ$), the experimental gap appears to deviate from the smooth and gradual dependence expected from the theory (inset to Fig. 2).

\cite{16} V. Melik-Alverdian, N.E. Bonesteel, and G. Ortiz, Bull. Am. Phys. Soc. 43, 496 (1998).

\cite{17} T. Jungwirth et al., to be published.

\cite{18} S. Melinte et al. (unpublished).
FIGURES

FIG. 1. $R_{xx}$ vs $B$ at $T=90$ mK and $\theta=0^\circ$. The vertical lines indicate some of the filling factors at which the QHE is observed. The inset shows the temperature dependence of $R_{xx}$ used to determine $\Delta$. From the slope of the least-square fit (dashed line), a gap of 20 K is obtained.

FIG. 2. $\Delta$ vs Zeeman energy, both in units of $e^2/\ell_B$. The corresponding tilt angles are indicated on the top axis. Inset: The energy gap to create a Skyrmion/Antiskyrmion pair (in units of $e^2/\ell_B$) as a function of $\tilde{g}$, from HF calculations with $w/l_B=0.52$ [12]. The vertical arrow indicates $\tilde{g}_c \approx 0.047$ above which a transition from Skyrmions to single spin-flip excitations is predicted. The dotted and dashed lines in the main figure and in the inset correspond to $K=9$ and $K=1$, respectively.

FIG. 3. $\xi=C/C_{QW}$ vs $\nu$ at $T=60$ mK at the indicated tilt angles. The absolute accuracy for $\xi$ ($\pm 15\%$) is shown. The $\xi$ envelope for $\theta=0^\circ$ (dotted line), is reproduced for comparison.

FIG. 4. $\xi$ vs $\nu$ at $T=60$ mK for $71^\circ \leq \theta \leq 74^\circ$. The dashed lines are guides to the eye.

FIG. 5. $\xi$ at $\nu>1$ (●) and $\nu<1$ (○) maxima is plotted vs $\tilde{g}$ at $\theta=0^\circ$, $\theta=46^\circ$ and $\theta \geq 71^\circ$.

FIG. 6. $\xi=C/C_{QW}$ vs $\nu$ at $T=60$ mK at the indicated tilt angles. The $\xi$ envelope for $\theta=0^\circ$ (dotted line), is reproduced for comparison.
Fig. 1
\[ \Delta / (e^2 / \varepsilon l_B) \]

Fig. 2

\[ \tilde{g} \equiv |g \mu_B B_{tot} / (e^2 / \varepsilon l_B) | \]
\[ \frac{\xi}{C/C_{QW}} \]

\[ \theta = 0^\circ \]

\[ \theta = 46^\circ \]

\[ \theta = 71^\circ \]

Fig. 3
Fig. 4

\[ \theta = 71^\circ \]

\[ \xi = \frac{C}{C_{Q W}} \]

\[ \theta = 72^\circ \]

\[ \theta = 73^\circ \]

\[ \theta = 74^\circ \]
\[ \xi = \frac{C}{C_{QW}} \]

\[ \bar{\xi} \equiv |g| \mu_B B_{tot} / (e^2/\epsilon^*_B) \]

Fig. 5
Fig. 6