Giant Low Temperature Heat Capacity of GaAs Quantum Wells near Landau Level Filling $\nu = 1$

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We report low temperature (T) heat capacity (C) data on a multiple-quantum-well GaAs/AlGaAs sample in the quantum Hall regime. Relative to its low field magnitude, C exhibits up to $\sim 10^5$-fold enhancement near $\nu = 1$ where Skyrmions are the ground state of the confined two-dimensional electrons. We attribute the large C to a Skyrmion-induced, strong coupling of the nuclear spin system to the lattice. The data are consistent with the Schottky nuclear heat capacity of Ga and As atoms in the quantum wells, except at very low T where C vs T exhibits a remarkably sharp peak, suggestive of a *phase transition in the electronic system*. [S0031-9007(96)00350-X]

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Heat capacity is one of the most fundamental physical properties as it directly probes thermodynamic quantities such as entropy [1]. In the case of two-dimensional electron systems (2DESs) heat capacity can be a powerful probe of single- and many-body properties such as the Landau quantized density of states and the quantum Hall effect (QHE), both integral and fractional [2,3]. Measurements of 2DES heat capacity, however, are among the most challenging experiments because of the very small electron contribution. As a result, in contrast to the overwhelming number of magnetotransport and magneto-optical experiments reported for 2DESs in the QHE regime [4], very few heat capacity measurements have been reported so far [5,6].

In this Letter we report heat capacity measurements of a modulation-doped GaAs/Al$_x$Ga$_{1-x}$As multiple-quantum-well heterostructure down to very low temperature ($T \geq$ 25 mK) and Landau level filling factor ($\nu \gtrsim 0.5$). As a function of increasing magnetic field, B, in addition to oscillations associated with the 2DESs' oscillating density of states at the Fermi level, we observe a dramatic increase of the low-T heat capacity (C) in the range $0.5 \lesssim \nu \lesssim 1.5$. For $0.7 \lesssim \nu \lesssim 0.85$ and $\nu \approx 1.2$, C exhibits a striking T dependence, including a remarkably sharp peak suggestive of a *phase transition* at very low T. We interpret these unexpected observations in terms of the Schottky model [7] for the nuclear-spin heat capacity of Ga and As atoms which couple to the lattice via the 2DESs' low energy excitation spin textures (Skyrmions). The origin of the peak at very low T is discussed in relation with a phase transition in the electronic system.

The experiments were performed on a multiple-quantum-well heterostructure grown by molecular-beam epitaxy and consisting of 100 GaAs quantum wells separated by Al$_{0.3}$Ga$_{0.7}$As barriers. The wells and barriers are 250 and 1850 Å thick, respectively, and the barriers are δ doped with donors (Si) near their centers. Electrical resistivity data on samples from the same wafer exhibit well-developed fractional QHE and attest to the high quality of the sample [8]. A 7×7 mm^2 piece of the wafer was thinned to 65 μm, and two carbon paint resistors were deposited on the substrate side and connected to the heat sink with NbTi wires. One carbon resistor was used as a thermometer while the other served as a heater. The carbon thermometer was calibrated versus a RuO$_2$ resistance thermometer at $B = 0$, and we checked that its calibration was negligibly affected by the magnetic field [9]. Depending on the external time constant of the sample τ_{ext}, three different techniques were used to measure C. At low B, $\tau_{\text{ext}} \approx 0.1$ s and C was measured using the ac technique [6,10] at 26 Hz, a frequency near which C was found to be frequency independent. In the B range near $\nu = 1$, C and hence τ_{ext} increased by up to 5 orders of magnitude, and we turned to a pulse technique [5,11] either in the relaxation regime ($C = \kappa \tau_{\text{ext}}$, where κ is the thermal conductance to the heat sink) or in the quasiadiabatic regime ($C = Q/\Delta T$, where ΔT is the temperature increase resulting from the applied heat Q after internal relaxation is completed), depending on τ_{ext}. C was measured in a dilution refrigerator while B was applied either perpendicular to the 2DES plane ($\theta = 0°$) or at an angle of $\theta = 30° \pm 2°$. The absolute accuracy on the measured C is of the order of $\pm(10\%-15\%)$, as illustrated in some of the figures.

The density of states at the Fermi level $D(E_F)$ of a 2DES in a perpendicular B exhibits $1/B$-periodic oscillations due to the formation of disorder-broadened Landau levels [2]. $D(E_F)$ is maximum at half-integral ν and is minimum at integral ν. The oscillating $D(E_F)$ induces oscillations in many physical properties such as electrical resistivity [2,4], magnetization [12], and electron specific heat: $c_e = (\pi^2/3)k_B^2 T D(E_F)$ (J/K) per electron [3]. The

data presented in Fig. 1(a) demonstrate the oscillatory behavior of C_e for a 2DES. Even though C is dominated by the lattice and addenda contributions, up to $\approx 10\%$ oscillations coming from the 2DESs are clearly observed. Comparison with previous data [6] gives $D(E_F)$ at $B = 2.3$ T ($\nu = 5/2$), about 5 times larger for our sample. This is consistent with the lower disorder, and hence smaller width of Landau levels, in our sample as evidenced by the presence of minima in C_e at odd ν down to $B = 1.14$ T ($\nu = 5$). From the position of the minima, we infer a 2DES density (per layer) of $n_e \approx 1.4 \times 10^{11}$ cm^{-2}, consistent with the magnetotransport data [8].

Near $\nu = 1$, C reveals a completely different and unexpected behavior. Figure 1(b) shows orders of magnitude enhancement of C with respect to the lower-B data of Fig. 1(a). C exhibits large maxima at $\nu \approx 0.8$ and 1.2 and decreases rapidly for $\nu \gtrsim 1.2$ and $\nu \lesssim 0.8$. Moreover, the T dependence of C is particularly striking (Fig. 2): In contrast to the low-B data where C *decreases* with decreasing T (not shown here), at $B = 7$ T ($\nu = 0.81$), C first *increases* with decreasing T and then displays a very sharp peak at $T_c \approx 36$ mK before decreasing at very low T [13]. In the remainder of the paper, we concentrate on the unexpected high-B data near $\nu = 1$.

We begin with a discussion of Fig. 2 data in the T range $0.1 \lesssim T \lesssim 0.4$ K, where $C \propto T^{-2}$. Both the very large magnitude of C and the T^{-2} dependence point to the nuclear Schottky effect which results from the

entropy reduction of the nuclear spin system (NSS) with decreasing T when the thermal energy $k_B T$ is much larger than the spin energy level spacing Δ [7]. The observation of the Schottky effect requires good coupling of the NSS to the lattice in order to reach thermal equilibrium in the time scale of the experiment. This coupling is provided by electron spin-flip excitations and further relaxation to the lattice [14]. Consequently, while the effect is commonly observed in metals, it usually remains undetected in high purity materials with low free-carrier density [15]. At first sight, because of their high purity and the low free-carrier density in the quantum wells, GaAs/AlGaAs heterostructures should not be good candidates for the observation of a nuclear Schottky effect. While this is supported by the low-B data [Fig. 1(a)] where only the lattice, addenda, and 2DESs contribute to the low value of C, surprisingly a Schottky behavior is observed at higher B near $\nu = 1$. Why?

Recent theoretical [16–18] and experimental [19–22] work on 2DESs has shown that, in the limit of a weak Zeeman coupling, electron spin textures known as *Skyrmions* are the lowest energy, charged excitations of the ferromagnetic ground state near $\nu = 1$ [2]. Skyrmions result from the dominance of the Hartree energy over the Zeeman energy, prohibiting single spin-flip excitations and favoring smooth distortions of the spin field; the competition between the two energies determines the total spin and size of Skyrmion quasiparticles.

While the importance of the electron-nuclear spin interaction has been known for some time [23,24], recent nuclear magnetic resonance (NMR) experiments [19,20] have provided clear evidence for a strong coupling of the NSS to the lattice through finite-size Skyrmions near $\nu = 1$. Subsequent theoretical results [18] have shown excellent quantitative agreement with the NMR data regarding the spin polarization of the 2DES, $\langle S_z \rangle$, thereby providing additional credence to the Skyrmionic picture
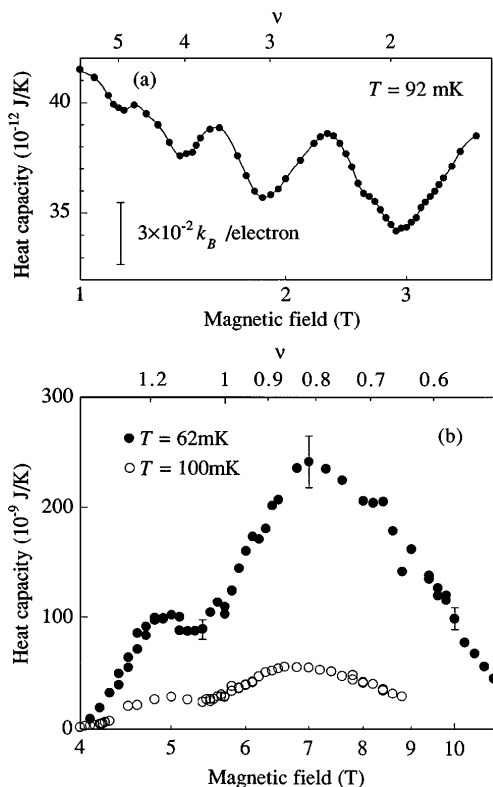
FIG. 1. Heat capacity C at $\theta = 0°$, showing orders of magnitude enhancement of the high-B data (b) over the low-B data (a). The line through the data points is a guide to the eye.
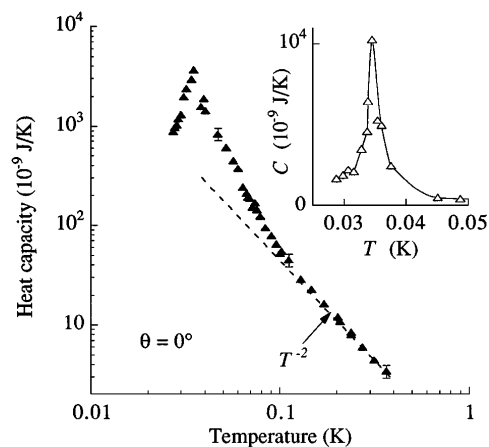
FIG. 2. The temperature dependence of C at $B = 7$ T ($\nu = 0.81$) is shown in the main figure in a log-log plot. The dashed line shows the T^{-2} dependence expected for the Schottky model. The inset shows a linear plot of C vs T at $B = 6.7$ T ($\nu = 0.85$).

near $\nu = 1$. Finally, very recently, further evidence for Skyrmions and estimation of their size was reported from magnetotransport [21] and magneto-optical data [22].

Based on these observations, we suggest that near $\nu = 1$ Skyrmions induce a strong coupling of the NSS to the lattice, and the large nuclear heat capacity is observed. The key role of Skyrmions is supported by (1) the absence of the nuclear-spin contribution to our measured C for $\nu \gtrsim 1.5$, where the Skyrmions are no longer the ground state of the 2DES [17,18,21], and (2) our experiments in a tilted B, presented in Fig. 3, which clearly show that the heat capacity anomaly relates to the 2DES filling factor (rather than total B).

We now turn to a more quantitative interpretation of the data. Ga and As have spin quantum number $I = 3/2$. When $\Delta \ll k_B T$, the (Schottky) nuclear specific heat is given by [1,25]

$$c_N = k_B \frac{5}{4} \left(\frac{\Delta}{k_B T} \right)^2 \left(\frac{J}{K \text{ nucleus}} \right), \qquad (1)$$

where $\Delta = \alpha B$, and $\alpha/k_B = 4.9 \times 10^{-4}$, 6.23×10^{-4}, and 3.5×10^{-4} K/T for ^{69}Ga (60.4%), ^{71}Ga (39.6%), and ^{75}As (100%), respectively [26]. If we assume that only the nuclear spins of the Ga and As atoms in the quantum wells contribute to the observed C, we obtain for our sample a nuclear heat capacity $C_N = 2.0 \times 10^{-11} B^2 T^{-2}$ (J/K) for $\Delta \ll k_B T$. Figure 3 clearly indicates that the calculated C_N is semiquantitatively consistent with both the size and the overall $\sim B^2$ dependence of the experimental data. The ratio of the experimental C and calculated C_N provides us with an estimate for the fraction (ξ) of Ga and As nuclei in the quantum wells that couple to the lattice. ξ shows maxima of the order of unity at $\nu \approx 0.85$ and 1.2, and decreases as $\nu \to 1$, and for $\nu \gtrsim 1.2$ and $\nu \lesssim 0.85$. While the decrease in ξ very near $\nu = 1$ can be attributed to the decreasing density of Skyrmions [18], its decrease very far from $\nu = 1$ (i.e., $\nu \gtrsim 1.2$ and $\nu \lesssim 0.85$) can be related to the 2DES approaching fillings where the

Skyrmions are no longer relevant. We note, moreover, that the ν dependences of C and ξ are qualitatively similar to that of $\langle S_z \rangle$ as deduced from the Knight-shift data [18,20]; in particular, both exhibit extrema at $\nu \approx 0.85$ and 1.2.

The temperature dependence of the high-B heat capacity at $\theta = 0°$ (Fig. 2) and $\theta = 30°$ (Fig. 4) at very low T is particularly striking. We observe that the $C \propto T^{-2}$ behavior is followed only down to ~ 0.1 K. For $T \lesssim 0.1$ K, C increases faster with decreasing T, and, in a narrow range of ν, C exhibits a remarkably sharp peak at very low T [13]. The deviation of C from the T^{-2} dependence at $T \gg \Delta/k_B$ and the shape of the observed peak are clearly not consistent with the Schottky model which predicts a smooth maximum in C at $T \sim \Delta/2k_B$ (~ 2 mK in our case). Instead, the shape and sharpness of this peak are suggestive of a *phase transition* [27]. The inset to Fig. 4 shows that the peak temperature T_c is maximum at $\nu \approx 0.8$ and 1.2, and decreases as $\nu \to 1$.

It is tempting to associate the sharp peak in C vs T observed at low T with the crystallization of Skyrmions, together with the associated magnetic ordering, near $\nu = 1$ which was recently proposed by Brey *et al.* [18]. The fact that our observed T_c decreases as $\nu \to 1$ is consistent with the decreasing Skyrmion density which should reduce the Skyrme-solid melting temperature. However, the details of the Skyrmion liquid-solid transition and, in particular, how it would affect the NSS are not known. Here we remark on possible interpretations of our data.

First, we note that Eq. (1) alone cannot account for the observed anomaly: The NMR data [20] suggest that Δ does not change significantly with decreasing T, and it is also expected [18,25] that a Skyrmion liquid-solid transition would affect Δ only very weakly. We might therefore interpret the substantial enhancement of C at low T as an indication that either (1) more nuclei couple to the lattice or (2) the entropy of the coupled NSS decreases faster with decreasing T than what is expected
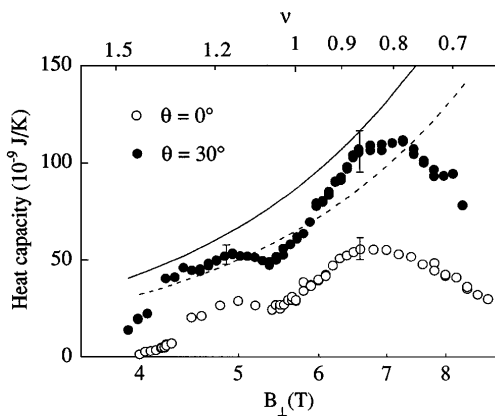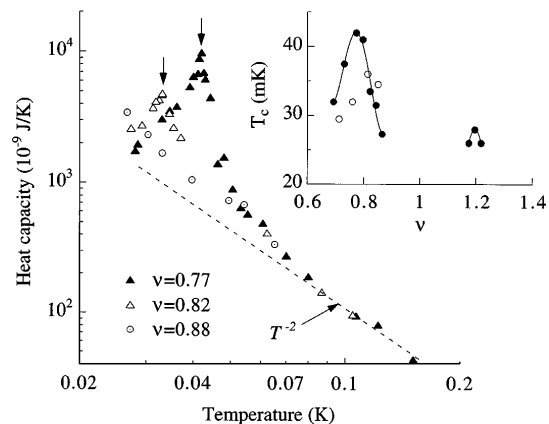

FIG. 3. Heat capacity as a function of perpendicular magnetic field $B_\perp = B \cos(\theta)$ and ν at $T = 100$ mK, and at the indicated values of θ. The curves correspond to the calculated nuclear-spin heat capacity (C_N) of Ga and As atoms in the quantum wells (C_N) for $\theta = 0°$ (dashed) and 30° (solid).

FIG. 4. Temperature dependence of heat capacity at $\theta = 30°$ and at the indicated values of ν. The T^{-2} dependence expected for the Schottky effect is shown as a dashed line, and T_c is marked by the vertical arrows. The ν dependence of T_c at $\theta = 0°$ ($\bigcirc$) and 30° ($\bullet$) is shown in the inset, and the lines are guides to the eye.

from the Schottky model. Picture (1) relies on a stronger coupling of the NSS to the lattice and also possibly on an enhanced nuclear spin diffusion so that a larger number of nuclei contribute to the heat capacity near T_c. This is consistent with T_c signaling the melting transition of Skyrmions: At such a phase transition, the coupling between the NSS and the electronic system is indeed expected to peak [25]. Picture (2), on the other hand, relies on a Skyrme-solid induced nuclear spin polarization which reduces the entropy of the NSS. This is reminiscent of the dynamic nuclear polarization of the NSS, for example, when nuclear spins interact with spin-polarized paramagnetic impurities [14]. While in the liquid Skyrme state motional narrowing prevents preferential orientation of electron spins, the transition to a pinned Skyrme solid could possibly induce a local preferential orientation of the electron-spin system, which in turn would polarize the NSS and thereby reduce the entropy. We emphasize that these are possible interpretations; a definitive conclusion regarding the origin of the observed low-T heat capacity anomaly awaits further experimental and theoretical work.

In conclusion, our heat capacity data underscore the importance of the coupling between the 2DES and nuclear spins, and point to the rich physics of the ground and excited states of the 2DES near $\nu = 1$.

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