# New Phases of the 2D Electron System in the Ultra-Quantum Limit Observed by Cyclotron Resonance

G. M. Summers,(1) R. J. Warburton,(1) J. G. Michels,(1) R. J. Nicholas,(1) J. J. Harris,(2)
and C. T. Foxon(3)
(1)*Physics Department, Clarendon Laboratory, Parks Road, Oxford, OX1 3PU, United Kingdom*
(2)*Semiconductor Interdisciplinary Research Center, Imperial College of Science and Technology,*
*Prince Consort Road, London, United Kingdom*
(3)*Physics Department, Nottingham University, Nottingham, United Kingdom*

(Received 11 November 1992)

Cyclotron resonance on low density, 2D electron systems in the ultra-quantum limit show split and shifted resonances. This demonstrates the presence in the ground state of both spin orientation electrons, whose relative proportion is strongly temperature and occupancy dependent. A critical occupancy of $\sim \frac{1}{10}$ divides single-particle behavior in a gaseous phase from the quantum liquid where the resonance positions are temperature and carrier density dependent. The resonances suggest a mixed spin state for some regions of the liquid phase.

PACS numbers: 73.20.Dx, 78.20.Ls

The phases of the two-dimensional electron system (2DES) in high magnetic field have been a subject of substantial interest recently [1-7]. Following the discovery of the fractional quantum Hall liquid (FQHL) states there is now very strong evidence for the existence of a Wigner solid (WS) state for both electron [1-4] and hole [5] systems. Suggestions have been made that both gas and liquid-crystal phases can exist at intermediate temperatures, or that domains of solid can persist up to temperatures well above the classical melting condition [6]. We report cyclotron resonance studies of the electron system in which we observe strong evidence for the existence of a phase boundary separating gaslike, independent single-particle behavior from a correlated liquid state, and we have clear evidence that at low quantum occupancy the liquid state is comprised of electrons of both spin states of the lowest Landau level, with a preference for spin pairing. By contrast the system appears to become completely spin polarized only at very low densities and low temperatures.

The experiment measures cyclotron resonance in a $^3$He cryostat with conventional infrared spectroscopy, using both a Fourier spectrometer and a $CO_2$ pumped molecular gas laser. A series of five high mobility GaAs/GaAlAs heterojunctions grown at Philips Research Laboratories, Redhill [7] was studied, with spacer layer thicknesses of 480, 320, 240, 160, and 80 nm, which allowed the carrier concentration to be varied from 1 to $15 \times 10^{10}$ cm$^{-2}$. In addition, continuous reduction of the density *in situ* by up to 80% was achieved using above barrier illumination from a HeNe laser with power densities of $\sim 10^{-7}$-$10^{-3}$ W/cm$^2$. No difference was detected between data taken at comparable densities either between measurements with different samples or with different illumination conditions. The carrier densities were measured to a precision of $\sim 10\%$, using the total absorption strength of the resonances. These values agreed well with transport data taken at low fields, although it was not usually possible to make electrical measurements on these samples at high magnetic fields. The magnetic field dependences were always measured *in the dark*, for which the density remained accurately constant, while carrier concentration studies were performed using fixed fields.

Figure 1 shows the transmission spectra for a series of magnetic fields for two electron densities, covering the range of occupancies $\nu = \frac{1}{5}$ to $\frac{1}{14}$ at 2.5 K. For higher occupancies a single cyclotron resonance is observed, but around $\frac{1}{6}$ a second resonance begins to appear on the higher energy side and the intensity progressively shifts over to this new peak while the total integrated intensity remains constant. Such behavior has been reported independently by two groups [8,9], and was initially interpreted in terms of formation of a Wigner solid. Nicholas *et al.* [8], however, pointed out that the magnitude of the splitting is very similar to that observed in bulk GaAs for the two different (spin conserving) cyclotron transitions

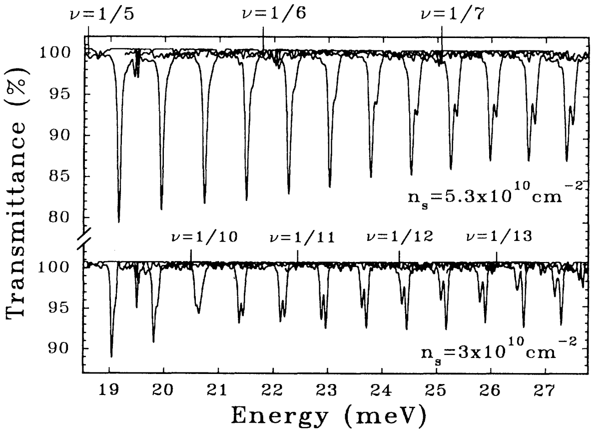

FIG. 1. A series of cyclotron resonance traces from 11.5 T to 17 T taken at 2.5 K for densities $3 \times 10^{10}$ cm$^{-2}$ and $5.3 \times 10^{10}$ cm$^{-2}$, covering the occupancy range $\frac{1}{5}$ to $\frac{1}{14}$.

© 1993 The American Physical Society

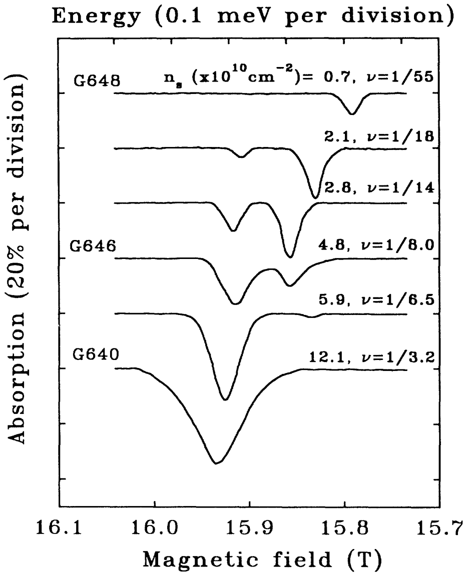

FIG. 2. The density dependence of the cyclotron resonance traces at 16 T and 2.2 K is shown covering the occupancy range $\frac{1}{50}$ to $\frac{1}{3}$. The three samples shown have spacer thicknesses 480, 240, and 120 nm. The ordering of the peaks is reversed in a field sweep compared with an energy sweep as shown in Fig. 1. The top axis shows an approximate equivalent energy scale.

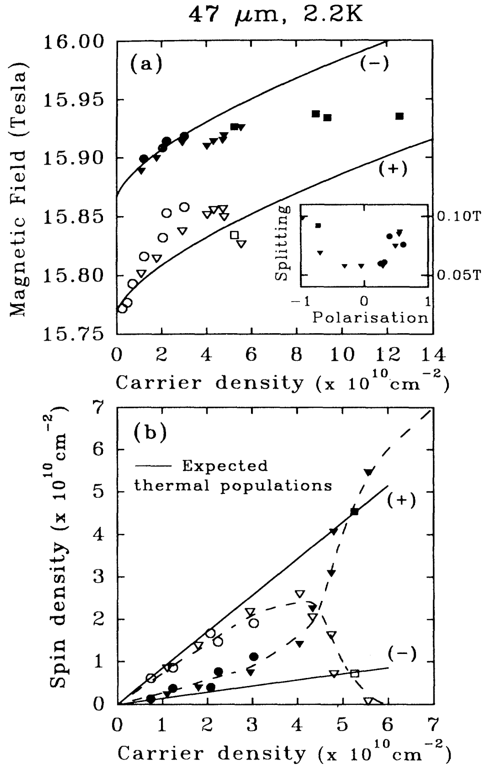

FIG. 3. (a) The position in magnetic field of the cyclotron resonance lines [open symbols spin (+), filled symbols spin (−)] is plotted against total carrier density for samples with spacer thicknesses 480 nm (circles), 240 nm (triangles), and 120 nm (squares). The solid lines show the expected behavior due to band nonparabolicity, calculated from the formula $m^*/m_e = 1 + 2.6\langle T_Z \rangle / E_g$, where $\langle T_Z \rangle$ is the kinetic energy due to the self-consistent quasiaccumulation layer potential [10]. The inset shows the anomalous decrease in splitting when the spin densities are equal. (b) The density per spin state is plotted against total carrier density. At low densities the populations follow the expected Boltzmann distribution (solid lines), calculated using the single-particle $g$ factor $-0.4$. The dashed lines are guides to the eye only.

originating from the spin (+) and spin (−) states of the lowest Landau level. The highest energy transition originates from the lowest energy spin (+) state due to the smaller spin splitting of the next Landau level caused by the energy dependence of the electron $g$ factor [10]. In the low density limit the absolute positions of both resonances correspond very accurately ($\mp 5$ mT) to the field positions in bulk GaAs, measured with the same system. The density dependent behavior of the resonances is shown in Fig. 2 near 16 T, where the occupancy is varied from $\sim \frac{1}{50}$ to $\frac{1}{3}$. At very low densities two resonances are observed with the higher energy (lower field) spin (+) resonance as the most intense and the second, weaker resonance at exactly the field expected for the spin (−) transition. As the carrier density increases there is a small continuous shift in the resonance positions shown in Fig. 3(a) due to the normal effect of band nonparabolicity. At around $\nu = \frac{1}{10}$ ($4 \times 10^{10}$ cm$^{-2}$) there is a rapid changeover in intensity between the two resonances and the resonance positions show an anomalous decrease in splitting, with the minimum corresponding to the point at which both intensities are equal. This can be seen in the inset, showing the splitting between the two peaks as a function of the polarization of the system defined as $(I_+ - I_-)/(I_+ + I_-)$, where the $I$'s represent the absorption strength in the two peaks. The majority of the shift seems to be associated with the nominally spin (+) transition. The carrier densities assigned to the spin (+) and spin (−) initial states, deduced from $I_+$ and $I_-$, are shown in Fig. 3(b) as a function of the total carrier density. The system shows a critical behavior with $I(+)$ and $I(-)$, swapping over at a critical occupancy ($\nu_c$) in the region of $\nu = \frac{1}{10}$. For the lowest densities, the carrier distribution is essentially given by the Boltzmann factor for the single-particle spin splitting [solid lines in Fig. 3(b)].

The existence of a critical occupancy ($\nu_c$) is confirmed by the temperature studies in Fig. 4. In the lower density case ($\nu = \frac{1}{12}$), as the temperature falls the intensity shifts rapidly to the transition corresponding to the lowest energy initial state [spin (+)], as expected for a single-particle system. Above $\nu_c$ the behavior is totally different. There is no observable change in the populations of the two states, but instead the two transitions change *position* continuously with temperature, and by 500 mK have merged to a single resonance at the *mean position.* Measurements at occupancies up to $\frac{1}{7}$ show that even

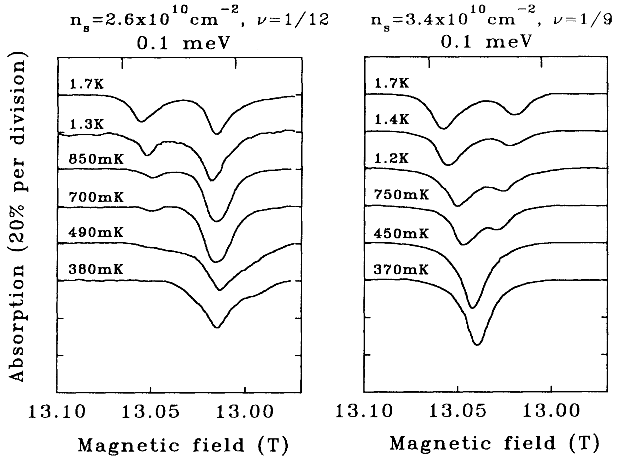

FIG. 4. Cyclotron resonance traces are shown as a function of temperature for occupancies $\frac{1}{12}$ and $\frac{1}{9}$ on either side of a critical value $\approx \frac{1}{10}$.

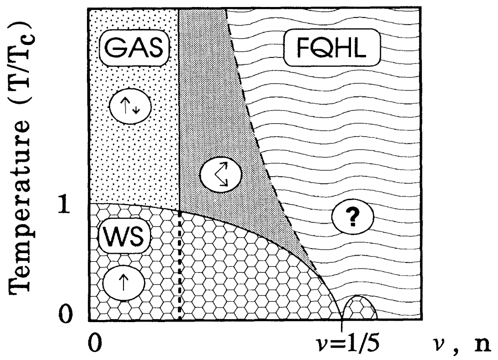

FIG. 5. A schematic phase diagram of the 2DES in the low occupancy region, showing the expected Wigner solid regime, the FQHL, the region of single-particle behavior, and the interacting doublet region. The circled symbols indicate the possible spin configuration of the system in each region.

when the initial resonance intensities are substantially different at higher temperatures (e.g., 2.5 K), the low-temperature limit of the resonance position is always close to the mean of the single-particle transitions. This also holds for occupancies up to $\frac{1}{3}$ where only a single peak is ever detected, but a temperature shift of the position is seen from 2 K to 300 mK which can be almost half of the single-particle spin splitting.

To understand this behavior we need to ask what the cyclotron resonance is measuring. Kohn's theorem [11] tells us that in the limit of a perfect system the long-wavelength cyclotron excitations come exclusively from the center-of-mass motion and are insensitive to electron-electron interactions. Under these conditions the resonance intensities will monitor the single-particle composition of the interacting multielectron ground state. Disorder effects are known to shift the cyclotron resonance positions and cause splittings [12] due to the removal of translational symmetry. However, it is unlikely that these could be responsible for the behavior seen here for three reasons. First, there is a systematic density dependence of the resonance intensities and positions which are continuous from one sample to the next, independent of the spacer layers, illumination states, and cooldown history. Second, the higher density resonance positions are strongly temperature dependent. This cannot be explained with a single-particle localized state picture. Finally, the low density limit of the resonance positions corresponds very accurately to the bulk values and the size of the splitting fits the known bulk spin splitting which has a characteristic $B^2$ dependence. This is very well fitted up to fields as high as 90 T, where carrier density dependent spin splittings have been observed at temperatures of 20 K and similar occupancies [13]. Kohn's theorem also breaks down when more than one carrier type is present with different masses. This may or may not lead to mixing of the different transitions, depending on the interactions present. Calculations by MacDonald and Kallin [14] suggest that electrons from two different Landau levels will mix and give a single excitation mode, but different spin states of the same Landau level will act independently at zero *k* vector. This is similar to the conclusions of earlier works [15], where it was shown that strong electron-electron interactions could mix even resonances from different subbands. Experimental searches [16] for the spin splitting at occupancies between 1 and 2 have shown that only a single transition can be observed in high purity GaAs/GaAlAs heterojunctions, to an accuracy of better than 10% of the expected splitting. The present results suggest that it is only in the extreme quantum limit that electron interaction effects are weak enough to allow both spin transitions to be observed.

The above data suggest that we can draw up a schematic phase diagram for the 2DES as shown in Fig. 5. At low temperatures and occupancies, WS formation occurs close to the classical WS melting temperature $T_c$ (typically 200–400 mK for the samples studied here) as suggested from the earlier work [1–6]. At low densities ($\nu < \frac{1}{10}$) and higher temperatures is the gas phase, where the carrier population is essentially single-particle-like. As the occupancy rises we enter the region where the FQHL forms, and our measurements show that this is a mixture of different spin states. A "doublet region" exists, approximately from $\nu = \frac{1}{10}$ to $\frac{1}{6}$, in which two resonances are observed whose intensities and positions are strongly temperature and occupancy dependent, but which should also form the WS at low temperatures (which our present results suggest may not always be completely spin polarized). Although the high density region ($\nu > \frac{1}{16}$) gives only a single peak, the temperature dependence of its position suggests that it is also not always a single-particle transition, but is a coupled resonance as seen for the integral occupancy region $1 < \nu < 2$ [16]. The strong temperature dependence of the resonance positions and the minimum in the splitting at equal spin occupancy suggest that the ground state may involve some spin pairing or realignment in the liquid phases. The low-temperature limit of the resonance positions corresponds to a transition energy to which there is no spin contribution, i.e., to either an $S=0$, an $S_z=0$, or a many-particle state in which the electron spins are aligned in the plane of the 2DES. The continuous nature of the transition with temperature suggests that a progressive spin realignment is taking place, with a canted spin alignment at intermediate temperatures. The formation of a mixed spin ground state is already well known for the case of certain FQHL states, specifically that at $\frac{2}{3}$, where it was both calculated [17] and shown experimentally [18] at very low fields that the ground state of the system could be spin unpolarized. It is also possible that even in the doublet region the second "spin $(-)$" resonance may not come from a pure spin state, but from one which is only partially polarized.

We now discuss possible origins of this new behavior. There is strong evidence for the existence of the WS phase at low temperature and calculations of the phase diagram [19,20] suggest that once the solid has melted, a region will exist in which short-range orientational order may persist as a hexatic liquid-crystal phase, up to a higher temperature ${}^2T_c$, above which no ordering remains. Electrical measurements are in general unable to access the very low occupancies studied here due to the formation of the insulating phase [1-3]. Luminescence measurements [6,21] report the observation of two critical temperatures, interpreted as due either to the presence of an intermediate hexatic phase [19,20] or a two-component mixture in which microcrystallites of solid coexist with liquid phase up to temperatures of order 2 K. The small domains of solid may be primarily the spin $(+)$ state thought to dominate the WS phase, while the liquid regions will give the hybridized resonances which dominate at higher densities. These pictures do not, however, include the spin degeneracy explicitly. This has been included recently in a calculation by Moulopoulos and Ashcroft [22] in which they suggest that the ground state of a 3D WS may be a paired electron crystal of $S=0$ spin-singlet pairs, due to the intrapair exchange forces, and that this will be more favored for a 2D system. Our results would seem to provide strong evidence that spin pairing occurs in 2D systems. More radical suggestions might include spin paired arrangements such as are seen in the textured states of liquid ${}^3$He [23] or as has been suggested for the high field vortex state of type-II 2D superconductors [24]. These would then involve the formation of triplet spin pairs, possibly leading to a preferential spin alignment in the plane of the 2DES.

In conclusion we may state that we have provided clear evidence for the existence of a gas-liquid phase boundary in the 2DES at high fields and for the important role played by spin degeneracy of the lowest Landau level. This leads to the observation of spin-split cyclotron resonance in the extreme quantum limit. We interpret the behavior of the resonance positions and amplitudes as evidence for a novel phase of the 2DES, in which spin pairing occurs.

[1] E. Y. Andrei *et al.*, Phys. Rev. Lett. **60**, 2765 (1988); D. C. Glattli *et al.*, Surf. Sci. **229**, 344 (1990).

[2] R. L. Willet *et al.*, Phys. Rev. B **38**, 7881 (1988).

[3] V. J. Goldman, J. E. Cunningham, M. Shayegan, and M. Santos, Phys. Rev. Lett. **65**, 2188 (1990); H. W. Jiang *et al.*, Phys. Rev. Lett. **65**, 633 (1990); F. I. B. Williams *et al.*, Phys. Rev. Lett. **66**, 3285 (1991).

[4] H. Buhmann *et al.*, Phys. Rev. Lett. **66**, 926 (1991); A. J. Tuberfield *et al.*, Surf. Sci. **263**, 1 (1992).

[5] M. B. Santos, Y. W. Suen, M. Shayegan, Y. P. Li, L. W. Engel, and D. C. Tsui, Phys. Rev. Lett. **68**, 1188 (1992).

[6] I. V. Kukushkin *et al.*, Phys. Rev. B **45**, 4532 (1992).

[7] C. T. Foxon, J. J. Harris, D. Hilton, J. Hewett, and C. Roberts, Semicond. Sci. Technol. **4**, 582 (1989).

[8] R. J. Nicholas *et al.*, in Proceedings of the Winterschool on New Developments in Low Dimensional Structures, Mauterndorf, 1992 (Springer-Verlag, Berlin, to be published).

[9] M. Besson, E. Gornik, C. M. Engelhardt, and G. Weimann, Semicond. Sci. Technol. **7**, 1274 (1992).

[10] H. Sigg *et al.*, Solid State Commun. **61**, 685 (1983); M. A. Hopkins *et al.*, Semicond. Sci. Technol. **2**, 568 (1987).

[11] W. Kohn, Phys. Rev. **123**, 1242 (1961).

[12] J. P. Cheng and B. D. McCombe, Phys. Rev. Lett. **64**, 3171 (1990).

[13] R. J. Nicholas, D. J. Barnes, N. Miura, J. J. Harris, and C. T. Foxon (to be published).

[14] A. H. MacDonald and C. Kallin, Phys. Rev. B **40**, 5795 (1989).

[15] J. Appel and A. W. Overhauser, Phys. Rev. B **18**, 758 (1978); C. S. Ting, S. C. Ying, and J. J. Quinn, Phys. Rev. B **16**, 5394 (1977).

[16] M. Watts, R. J. Nicholas, N. J. Pulsford, J. J. Harris, and C. T. Foxon, in *Proceedings of the Twentieth International Conference on the Physics of Semiconductors,* edited by E. M. Anastassakis and J. D. Joannopoulos (World Scientific, Singapore, 1991), p. 1465.

[17] T. P. Chakraborty, P. Pietalainen, and F. C. Zhang, Phys. Rev. Lett. **57**, 130 (1986); Phys. Rev. B **30**, 7320 (1984).

[18] J. P. Eisenstein, H. L. Stormer, L. N. Pfeiffer, and K. W. West, Phys. Rev. B **41**, 7910 (1990); R. G. Clark, in *Localization and Confinement of Electrons in Semiconductors,* edited by F. Kuchar, H. Heinrich, and G. Bauer (Springer, Berlin, 1990), p. 168.

[19] B. I. Halperin and D. R. Nelson, Phys. Rev. Lett. **41**, 121 (1978); Phys. Rev. B **19**, 2457 (1979); A. P. Young, Phys. Rev. B **19**, 1855 (1979); for a general review of 2D melting, see K. J. Strandburg, Rev. Mod. Phys. **60**, 161 (1988).

[20] C. A. Murray and D. H. van Winkle, Phys. Rev. Lett. **58**, 1200 (1987).

[21] E. M. Goldys *et al.*, Phys. Rev. B **46**, 7957 (1992).

[22] K. Moulopoulos and N. W. Ashcroft, Phys. Rev. Lett. **69**, 2555 (1992).

[23] A. J. Leggett, Rev. Mod. Phys. **47**, 331 (1975).

[24] H. Akera, A. H. MacDonald, S. M. Girvin, and M. R. Norman, Phys. Rev. Lett. **67**, 2375 (1991); Phys. Rev. B **45**, 10147 (1992).