Thermodynamics of melting of a two-dimensional Wigner electron crystal

1991 J. Phys.: Condens. Matter 3 4301

(http://iopscience.iop.org/0953-8984/3/23/020)

J. Phys.: Condens. Matter 3 (1991) 4301–4306. Printed in the UK

LETTER TO THE EDITOR

Thermodynamics of melting of a two-dimensional Wigner electron crystal

M J Lea†, N H March‡ and W Sung‡§

† Department of Physics, Royal Holloway and Bedford New College, University of London, Egham, Surrey TW20 0EX, UK

‡ Theoretical Chemistry Department, University of Oxford, South Parks Rd., Oxford OX1 3UB, UK

Received 28 March 1991

Abstract. The thermodynamics of the Wigner crystallization of two-dimensional electrons in a magnetic field is set out. The results relate the slope of the melting curve as a function of the Landau level filling factor ν, to changes in magnetization, ΔM, and entropy, ΔS, on melting, and identify points where ΔM, ΔS and $\Delta E = 0$, with E being the internal energy. The phase diagram as revealed by recent experiments on GaAs/AlGaAs heterojunctions is thereby analysed. Remarkable features are inferred in the magnetization of the strongly correlated electron liquid.

The proposal made by Durkan *et al* (1968) that localization of electrons in impure semiconductors by strong magnetic fields could assist Wigner crystallization has been brought to fruition by Andrei *et al* (1988), Glattli *et al* (1990) and Jiang *et al* (1990). These workers used the two-dimensional electron gas (2DEG) in GaAs/AlGaAs heterojunctions in strong magnetic fields to observe the magnetically-induced Wigner solid (MIWS). Their major findings have been confirmed by Buhmann *et al* (1991) who observed specifically an additional line in the luminescence spectrum of such a heterojunction below a critical Landau level filling factor ν_c and a critical temperature ($T_c = 1.4$ K at 26 T). The lack of correlation between ν_c and the disorder-related properties of the system testifies to the intrinsic nature of the line and its appearance, they conclude, signals the formation of a Wigner solid.

In the above experiments, it proved possible to map out approximately the melting curve of the Wigner solid as a function of the Landau level filling factor, ν, given in terms of the (areal) electron density n and the magnetic field H applied perpendicular to the electron layer, by

$$\nu = nhc/eH. \tag{1}$$

The schematic phase diagram proposed by Buhmann *et al* (1991) is shown in figure 1. There are four crystal phases, marked C1, C2, C3 and C4, interspersed with the liquid phase at filling factors corresponding to the fractional quantum Hall effect (FQHE) at $\nu = \nu_q = 1/q$ with $q = 5$, 7 and 9. The final solid phase ends at a critical filling factor of $\nu_c = 0.28 \pm 0.02$. These experimental results have prompted us to generalise the

§ Permanent address: Department of Physics, Pohang Institute of Science and Technology, Korea.

0953-8984/91/234301 + 06 $03.50

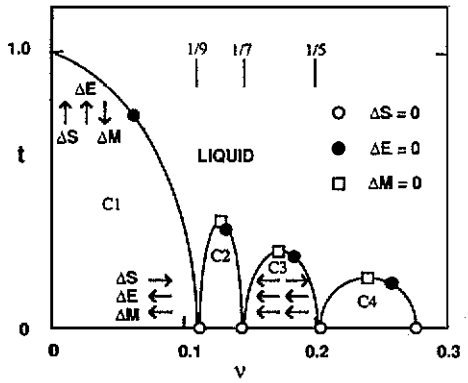

Figure 1. Schematic phase diagram as proposed by Buhmann *et al* (1991), showing the four crystal phases, C1 to C4, and the reentrant liquid phase at $\nu = \frac{1}{5}$, $\frac{1}{7}$ and $\frac{1}{9}$. The symbols mark the points ΔM, ΔS and $\Delta E = 0$ as deduced from thermodynamics. The arrows show the direction of ΔM, ΔS and ΔE across the phase boundary nearest to the arrows. The arrows in phases C2 and C4 match those shown in phase C3. It should be noted that, as alternatives, the possibilities of a finite liquid range at $T = 0$ or a solid always stable at the lowest temperatures, are not excluded.

thermodynamics of Wigner crystallization, already considered by Parrinello and March (1976) in zero magnetic field. The starting point of the present study of the thermodynamics of an electron crystal to electron liquid first-order melting transition is the result (see, for example, Pippard 1966) for the melting temperature, T_m, as a function of magnetic field, H. At constant area Ω, this can be written

$$(\partial T_m/\partial H)_\Omega = -\Delta M/\Delta S. \tag{2}$$

If the subscript C denotes the crystal phase and the subscript L the liquid phase, then $\Delta M = M_L - M_C$ is the change of magnetization on melting, while $\Delta S = S_L - S_C$ is the corresponding entropy change. This equation is readily cast into a form directly useful in analysing the measurements referred to above by using the relation between ν and H given in (1) to find

$$(\partial T_m/\partial \nu)_\Omega = (H/\nu)\,\Delta M/\Delta S. \tag{3}$$

Let us comment immediately on some properties which follow from (3):

(i) Turning points in the melting curve in the (ν, T) plane correspond to $\Delta M = 0$, provided the entropy change, ΔS, does not simultaneously go to zero. This result means that maxima (see figure 1) in the melting curve immediately locate points where the magnetization in the strongly correlated liquid equals that in the Wigner solid.

(ii) Infinite slopes on the melting curve, at specific values of ν away from $\nu = 0$, imply $\Delta S = 0$ for non-zero ΔM.

(iii) (3) can also be used to locate points where $\Delta E = 0$, where E is the internal energy. If we write the magnetic Helmholtz free energy, F, relevant to phase equilibrium at constant T, field H and area Ω, as (Pippard 1966)

$$F = E - TS - HM \tag{4}$$

the equilibrium condition $F_L = F_C$ evidently then yields

$$\Delta E - T\Delta S - H\Delta M = 0. \tag{5}$$

As envisaged in (iii) above, the existence of a point, or points, on the melting curve where $\Delta E = 0$ yields at such points

$$\Delta M/\Delta S = -T_m/H \tag{6}$$

the slope of the melting curve is then given by (from (3))

$$(\partial T_m/\partial \nu)_\Omega = -T_m/\nu. \tag{7}$$

This equation can only be satisfied for $\partial T_m/\partial \nu$ negative and then whenever the melting

curve lies at a tangent to a hyperbola of the form $T_m\nu$ = constant. Hence we now have a prescription for locating points where ΔM, ΔS and ΔE are zero, and these have been marked on the phase diagram in figure 1. At each point the relative signs of the non-zero differentials are then fixed by the relations in (5). Also, equation (5) requires that ΔS and ΔM cannot simultaneously have opposite signs to ΔE.

Following Parrinello and March (1976), it is also useful to invoke the virial theorem in this treatment of Wigner crystallization. For non-zero magnetic field this reads

$$2K + 2HM + U = 2p\Omega \tag{8}$$

where K is the kinetic energy, U potential energy ($E = U + K$) and p the pressure. Equation (8) is for a two-dimensional electron assembly with a three-dimensional Coulomb interaction (Tao 1990). For incompressible phases not affected by pressure we have†

$$2\Delta K + 2H\,\Delta M + \Delta U = 0. \tag{9}$$

Combining this with (5) we identify two additional points $\Delta U = 0$ and $\Delta K = 0$, which occur when $\partial T_m/\partial \nu = -T_m/2\nu$ and $-T_m/3\nu$, respectively. In summary we have

(i) When $\Delta M = 0$,

$$\Delta E = T_m\,\Delta S = \tfrac{1}{2}\Delta U = -\Delta K. \tag{10}$$

(ii) When $\Delta S = 0$ or $T = 0$,

$$\Delta E = H\,\Delta M = \tfrac{1}{4}\Delta U = -\tfrac{1}{3}\Delta K. \tag{11}$$

(iii) When $\Delta E = 0$

$$T_m\,\Delta S = -H\,\Delta M = -\tfrac{1}{2}\Delta U = \tfrac{1}{2}\Delta K. \tag{12}$$

(iv) When $\Delta U = 0$

$$\Delta E = -H\,\Delta M = \tfrac{1}{2}T_m\,\Delta S = \Delta K. \tag{13}$$

(v) When $\Delta K = 0$,

$$\Delta E = -2H\,\Delta M = \tfrac{2}{3}T_m\,\Delta S = \Delta U. \tag{14}$$

Let us now consider some of these results in the light of the schematic phase diagram (figure 1) from Buhmann *et al* (1991), into which they have subsumed the main features compatible with microscopic theory and experiment. It is clear (see Andrei *et al* 1988) that, as ν tends to zero, the melting temperature, T_m, should tend to that of a classical one-component plasma. This has a melting temperature, T_{mc}, given by

$$T_{mc} = e^2(\pi n)^{1/2}/\kappa k_B \Gamma_m \tag{15}$$

where k_B is Boltzmann's constant while $\Gamma_m = 127 \pm 3$ (Deville 1988) and κ is the dielectric constant of the host material. Hence figure 1 shows $t_m = T_m/T_{mc}$ versus ν with $t_m = 1$ at $\nu = 0$. As this point is approached, we anticipate on physical grounds that the entropy S_L of the liquid will be greater than that of the solid S_C‡. Since there is no physical reason for an infinite slope of the melting curve at $\nu = 0$, it is clear from (1) and (3) that, with

† Fully quantitative work may require an estimate of the electron pressure contribution relative to the magnetic field term.

‡ Should this expectation not be borne out by experiment, the changes to be made in figure 1 are straightforward.

ΔS not equal to zero, ΔM must approach zero at least as rapidly as ν^2 along the melting curve, at constant density.

We have labelled figure 1 with the points corresponding to $\Delta M = 0$, $\Delta E = 0$ and $\Delta S = 0$. The arrows show the directions in which S, M and E increase across the melting curve. For phase C1, the arrows are completely determined by the condition $\Delta S > 0$ for the classical crystal at $\nu = 0$, which also implies $\Delta M < 0$ and $\Delta E > 0$ in the same limit. In this picture $\Delta S = 0$ occurs only at $T = 0$ as required by the third law of thermodynamics, which implies that the melting curve approaches the axis with a vertical slope. The arrows in the other regions C2, C3 and C4 follow from either (i) immediately above, taking $\Delta S > 0$ at the highest temperatures in each solid phase, or (ii) assuming that the thermodynamic functions behave in a similar way each time the phase diagram approaches $\nu = 0$. Figure 2 shows the points at which $\Delta E = 0$, $\Delta U = 0$ and $\Delta K = 0$ that occur for $\partial t_m/\partial \nu = -t_m/\nu$, $-t_m/2\nu$ and $-t_m/3\nu$, respectively.

The simplest conceptual picture that emerges from this phase diagram is that the four separate solid-phase regions are essentially the same Wigner solid (presumably a triangular crystal with one electron per lattice point on a 2D hexagonal Bravais lattice) with regions near $\nu = \frac{1}{5}$, $\frac{1}{7}$ and $\frac{1}{9}$ where the liquid phase has a lower free energy. It is conceivable that the four solid phases are different but this seems somewhat unlikely, particularly since the ν values indicate that the change in ordering is occurring in the liquid. There have been many theoretical calculations of the ground state energies of the Wigner solid and the fractional quantum Hall effect liquid states (Levesque *et al* 1984, Lam and Girvin 1984, Yoshioka *et al* 1983, Halperin 1984, Egorov 1986, Markiewicz 1986, Thugman and Kivelson 1985, Kivelson *et al* 1987, Isihara 1989). There is a consensus that the solid has lower energy at very small values of ν but that the FQHE states with $\nu = 1/q$, as introduced by Laughlin (1983), have lower energies above some critical $\nu_c \sim 1/6.5$ (Lam and Girvin 1984). The energy of the FQHE states also has cusps at $\nu = 1/q$ with the limiting slopes being related to the energy gap of the quasiparticle excitations in the liquid (Halperin *et al* 1984).

Figure 1 shows that the entropy change on melting, ΔS, is greater than zero on either side of the liquid state at ν_q. We assume that, at low temperatures,

$$\Delta S = A(\nu)T^{\alpha} \tag{16}$$

where $A(\nu)$ and α are positive constants which may vary strongly around ν_q. (The entropy from the thermal excitations from the ground state at ν_q of the form $\exp(-\Delta/kT)$, where Δ is the quasiparticle energy gap, will be negligible near $T = 0$.) This entropy could come from low-lying excitations in the liquid or from quasiparticle excitations related to $|\nu - \nu_q|$ (which might give $\alpha = 0$). Then, on the melting curve†,

$$T_m \Delta S = \Delta E - H\Delta M = \Delta \mathcal{G} = A(\nu)T_m^{\alpha+1}. \tag{17}$$

The magnetization of the liquid, M_L, will be discontinuous at a cusp in the energy at $\nu = 1/q$, and could change sign (see Isihara 1989). If the change in magnetization on melting ΔM is approximately equal to M_L, then the relative signs of ΔM agree with the conclusions from figure 1, as sketched in figure 3, assuming that the transitions occur each side of ν_q. This field dependence of ΔM is very reminiscent of the de Haas–van Alphen effect at integral ν values, suggesting that the magnetism of the electron liquid phase is intimately connected with the exotic variation of ΔM shown here. The conclusion here

† $\Delta\mathcal{G}$, the free energy at $T = 0$, has the re-entrant cusp minima following the melting curve (figure 1). Since $\mathcal{G}$, is a monotonically increasing function of ν, this feature is entirely due to the electron liquid, consistent with the theoretical results mentioned earlier.

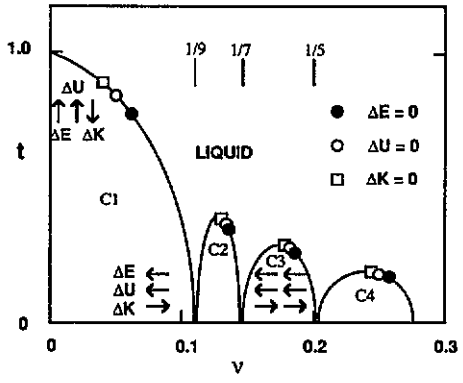

Figure 2. Schematic phase diagram showing the points ΔE, ΔU and $\Delta K = 0$ as deduced from thermodynamics. The arrows show the direction of ΔE, ΔU and ΔK across the phase boundary nearest to the arrows. The arrows in phases C2 and C4 match those shown in phase C3.

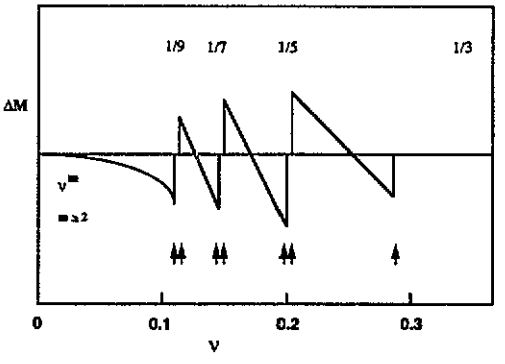

Figure 3. Schematic diagram of the change in magnetization on melting along the melting curve shown in figure 1. The arrows mark the liquid–solid transitions.

is that, for a finite width of reentrant liquid phase at finite temperature, the magnetization and entropy are properties of the liquid, connected by (17) on the melting line. However as T decreases towards zero, the entropy must go to zero. If this occurs by some further ordering among the excitations, then ΔM may also go to zero at the same time. If $\Delta M/\Delta S$ remained a constant in this process then $\partial t_m/\partial \nu$ would approach the axis with a finite slope, from (3). While it is not inconceivable that the orbital magnetism of the strongly correlated electron liquid could be consistent with the anyon model (Wilczek 1990), we are currently studying simpler microscopic models. The presence of minima in the free energy could also lead to the possibility of other phenomena, such as phase separation. However the relative energies of the different phases are small compared with the total Coulomb energy of the electron sheet so that the free energy versus density plot will probably exhibit only a small cusp with no minima at ν_q.

One of us (WS) wishes to acknowledge support from Pohang Institute of Science and Technology and the Ministry of Education in Korea during his sabbatical leave in Oxford.

References

Andrei E Y, Deville G, Glattli D C, Williams F I B, Paris E and Etienne B 1988 *Phys. Rev. Lett.* **60** 2765
Buhmann H, Joss W, von Klitzing K, Kukuskin I V, Plaut A S, Martinez G, Ploog K and Timofeev V B 1991 *Phys. Rev. Lett.* **66** 926
Deville G 1988 *J. Low Temp. Phys.* **72** 135
Durkan J, Elliott R J and March N H 1968 *Rev. Mod. Phys.* **40** 812
Glattli D C, Deville G, Duburcq G, Williams F I B, Paris E and Etienne B 1990 *Surf. Sci.* **229** 334
Egorov V S 1986 *Sov. Phys.–Solid State* **28** 177
Isihara A 1989 *Solid State Physics* vol 42 (New York: Academic) p 271
Jiang H W, Willett R L, Stormer H L, Tsui D C, Pfeiffer L N and West K W 1990 *Phys. Rev. Lett.* **65** 633
Kivelson S, Kallin C, Arovas D A and Schrieffer J R 1987 *Phys. Rev.* B **36** 1620
Halperin B I 1984 *Phys. Rev. Lett.* **52** 1583
Lam P K and Girvin S M 1984 *Phys. Rev.* B **30** 493

Laughlin R B 1983 *Phys. Rev. Lett.* **50** 1395
Levesque D, Weis J J and McDonald A H 1984 *Phys. Rev.* B **30** 1056
Markiewicz R S 1986 *Phys. Rev.* B **34** 4172; *Phys. Rev.* B **34** 4177
Parrinello M and March N H 1976 *J. Phys. C: Solid State Phys.* **9** L147
Pippard A B 1966 *Elements of Classical Thermodynamics* (Cambridge: Cambridge University Press)
Tao Z C 1990 *Phys. Lett.* **151A** 172
Thugman S A and Kivelson S 1985 *Phys. Rev.* B **31** 5280
Wilczek F 1990 *Anyons* (Singapore: World Scientific) p 87
Yoshioka D, Halperin B I and Lee P A 1983 *Phys. Rev. Lett.* **50** 1219