Thermodynamic Measurement on the Melting of a Two-Dimensional Electron Solid

D. C. Glattli, E. Y. Andrei,[a] and F. I. B. Williams
Service de Physique du Solide et de Résonance Magnétique, Centre d'Etudes Nucléaires de Saclay, 91191 Gif-sur-Yvette Cedex, France
(Received 9 November 1987)

Absolute specific-heat measurements are presented for the two-dimensional Coulomb solid phase and an upper limit of $0.2k_B$ per particle is established for the change in entropy on melting. The results support the dislocation-pair–unbinding model of Kosterlitz and Thouless.

PACS numbers: 64.70.Dv, 65.40.Em, 67.70.+n

The solid-liquid phase transition in two dimensions has proved to be a less simple problem than was once imagined. Even the simplest example of particles in a uniform two-dimensional (2D) space with repulsive power-law interaction is argued to show different melting behavior according to the relative energies of the defects involved. The first and attractively simple idea proposed by Kosterlitz and Thouless[1] was that such systems might melt because of the basic instability of a solid to the destruction of rigidity which accompanies the thermodynamic dissociation of dislocation pairs. But this instability only sets an upper bound to the temperature at which a solid may exist. For example, Chui[2] pointed out that it is possible for dislocations in the solid to group themselves into grain boundaries, in which case the melting can occur at lower temperature and become of first order. Density-wave theories[3] also suggest preempting of the dislocation instability, again with the first-order behavior of the thermodynamic potential.

Indeed 2D melting does not seem to have a single universal behavior. For example, numerical experiments on Lennard-Jones systems all give first-order melting[4] whereas laboratory experiments show it to be continuous in certain cases.[5] But one can argue that computations are limited in time and size while experiments are complicated by substrate structure. For power-law interactions, most numerical work indicates first-order melting while the laboratory experiments are more in favor of continuous-order transitions. It is of interest then to take a very "clean" experimental system and do a series of experiments designed specifically for the understanding of melting.

Electrons deposited on the vapor side of a liquid-helium surface are not only the physically realized system closest to the theoretical model, but they also are relatively easy to probe. Perpendicular motion is frozen into an isolated singlet quantum level while parallel (2D) motion is classical in the sense that $T \gg T_F$ and is unrestricted except by small random potentials due to helium surface excitations which have time average zero. The interaction potential, which is pure Coulomb $1/r$ up to a screening length of several hundred interparticle spacings, induces a liquid-solid transition as the temperature is lowered.[6] Experiments have been done at melting[7] which show that the shear modulus is within 10% of the value required for stability of the solid phase to dislocation-pair dissociation. If this is indeed the cause of melting, the thermodynamic potential is expected to have only an essential singularity (continuous transition). The present experiment is aimed at giving a direct answer about the order of the transition by making a thermodynamic measurement of the specific heat.

Specific heat of solid phase.— We seek the heat capacity of $\simeq 10^8$ electrons on a sea of $\simeq 10^{22}$ helium atoms. It is quite evidently futile to make a global measurement. But because the electrons are coupled to the helium thermostat by scattering off the more massive capillary waves of the liquid helium, they only lose a fraction of their energy at each collision so that the thermal (energy) relaxation times $\tau_{e\text{-He}}$ can be considerably longer than internal electron-electron equilibrium times $\tau_{e\text{-}e}$. It is then possible to define an electronic temperature T_e different from the bath temperature T_b. Then if we heat the electrons with a pulse of power P for a time $\tau_{e\text{-}e} \ll \Delta t \ll \tau_{e\text{-He}}$, we can directly determine the specific heat C by monitoring the electron temperature variation ΔT_e:

$$C = \Delta Q/\Delta T_e = P\Delta t/\Delta T_e, \tag{1}$$

without interference from the large helium reservoir.

Experimentally the electrons are confined on the liquid-helium surface of a half-filled, circular, plane parallel capacitor (radius $R = 11$ mm, separation $D = 2$ mm). The top electrode is held at a static potential $-|V|$ and the cylindrical guard ring at $-|W| < -|V|$ with respect to the bottom electrode. Low holding potentials $|V| < 4\pi n_0 eD$ and low densities $n_0 < 2\times 10^8$ cm^{-2} are used in order to increase the thermal relaxation time $\tau_{e\text{-He}}$. Lying on the bottom electrode, a 50-Ω meander transmission line similar to that used by Deville and co-workers[7,8] creates a plane-wave potential in the electron plane which can excite the low-wave-vector longitudinal modes of the electron disk. A broad-band radio-frequency spectrometer (1–500 MHz) detects the resonant excitation frequencies by the loss in the power transmitted by the line. In the liquid

phase these are the low-wave-vector longitudinal modes (plasmons) of frequency

$$\omega_p(k_{\nu,\mu}) = c_p k_{\nu,\mu}, \tag{2}$$

where $c_p = [2\pi n_0 e^2 D/(1+\epsilon)m]^{1/2}$ is the screened plasmon velocity, ϵ the liquid-helium dielectric constant. In the solid phase, the frequencies are modified to

$$\omega(k) = [\omega_p^2(k) + \omega_0^2(T_e)]^{1/2}. \tag{3}$$

The boundary conditions on the disk perimeter quantize the wave number k through $J_\nu'(k_{\nu,\mu}R) = 0$.[8] For small electron-ripplon coupling, the first ten modes are well resolved. One of them is used to heat the electrons and another to measure the electron temperature. The thermometry is based on the electron-temperature dependence of the ω_0 term in (3) which results from the electrons, localized in their own lattice, being pushed onto the helium surface by the holding field and imprinting a lattice of small dimples in which they vibrate at the frequency ω_0.[9] The dimple depth, and so ω_0, are direct functions of the spatial extent of the electron fluctuations $\langle u^2 \rangle$ averaged over the slow ripplon time scale. As $\langle u^2 \rangle \sim T_e$, the ω_0 variations give a direct measure of the electron temperature independent of the helium temperature.

A low-level rf source G_1 monitors one of these lowest modes whose frequency is sensitive to T_e at a frequency f_1 corresponding to a point of greatest slope on the resonance absorption line. The change in the transmitted power is then proportional to the electron temperature variation. As we can calibrate the thermometer against the helium temperature under equilibrium conditions where $T_e = T_b$, this signal is used to make an absolute measurement of the electron temperature.

The electrons are heated by our pulsing a second generator G_2 on resonance with another mode at frequency $f_2 = \omega_{\nu,\mu}/2\pi \gg f_1$. Of this power $P_0 < 0.1$ nW a small fraction is absorbed by the electrons:

$$P = Ne^2 |E_{\nu,\mu}|^2 \tau/2m, \tag{4}$$

where $E_{\nu,\mu}$ is an effective electric field calculable from P_0 and the geometry and $\tau = (\Delta\omega)^{-1} \gtrsim 10^{-8}$ s is given by the linewidth $\Delta\omega$.

Figure 1 shows the time evolution of the electron temperature during and after the pulse. For the entire range of equilibrium temperatures T_b explored, and for small electron temperature deviations, an exponential relaxation is found with time constant $2\times10^{-5} > \tau_{e\text{-He}} > 10^{-5}$ s. By continuous heating we find an electron-bath thermal conductivity K defined by $P = K(T_e - T_b)$ consistent with the exponential law:

$$C(dT_e/dt) = -K[T_e(t) - T_b], \quad C/K = \tau_{e\text{-He}}. \tag{5}$$

This relaxation time is long enough for internal equilibrium to be achieved.

Absolute specific-heat values in the solid phase are given in Fig. 2. They are obtained through relation (1) or (5). As a check, we increased the pressing field to reduce $\tau_{e\text{-He}}$ by a factor of 2 and verified that the values of C are unaltered. They compare well with a pure phonon heat capacity calculated from the zero-temperature dispersion relations[10] with the value $c_t(0)$ of the transverse sound-velocity phonon renormalized to $c_t^2(T) = c_t(0)^2(1 - \gamma T/T_m)$, where $\gamma = 0.3$ has been measured previously.[7] It is because of the low Debye temperature $T_D = 0.935 T_m (n_0/10^8\ \text{cm}^{-2})$ that the values are rather

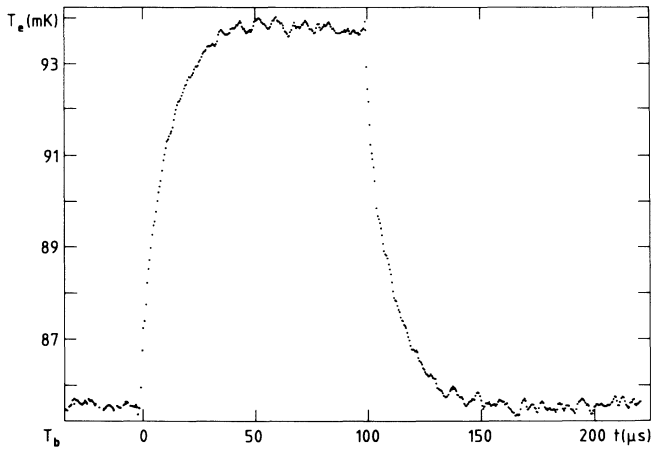

FIG. 1. Average of $\simeq 10^5$ recordings of electron temperature (T_e) induced by heating pulse (P) for a solid of density $n = 1.02\times10^8$ cm^{-2} at a helium temperature $T_b = 84.5$ mK and holding electric field 110 V cm^{-1}.

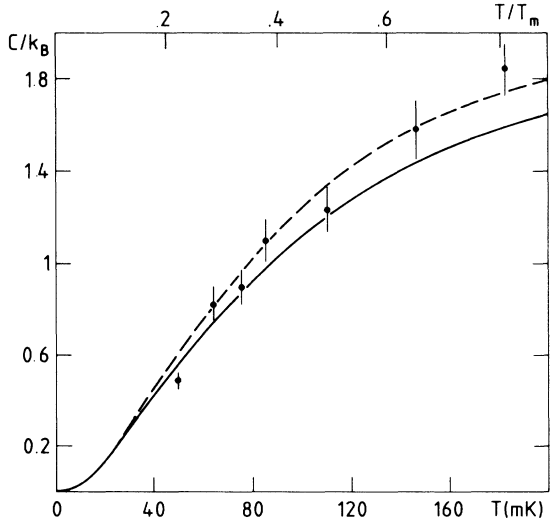

FIG. 2. Specific heat per electron in solid phase as a function of reduced temperature T/T_m. Experimental values are compared with the phonon specific heat [solid line, $c_t = c_t(0)$; dashed line, $c_t(T) = c_t(0)(1 - 0.3T/T_m)$].

close to the classical limit of $2k_B$ and the low-temperature limit $\sim T^2$ is not attained. At 20% below T_m no defect contribution is discernible; this is consistent with a theoretical estimate of $0.1k_B$ from the results of Fisher, Halperin, and Morf.[11]

Latent heat of melting.—As the thermometer variable vanishes at melting, we modified the experiment by coupling the electron sample under test, of density n_1 and melting temperature T_{m1}, to a second sample of higher density n_2 which remains solid ($T_{m2} > T_{m1}$) and whose nonvanishing local-mode frequency $\omega_0^{(2)}(T_e)$ affords us a thermometer. The latter can be regarded as the calorimeter and the former as the sample. If thermal equilibrium between calorimeter and sample is reached well before equilibrium with the helium bath ($K_{12} \gg K_{1\text{-He}}$, $K_{2\text{-He}}$; see Fig. 3), then by heating the calorimeter and recording its temperature T_2 ($=T_1$) we can deduce the total specific heat $C = C_1 + C_2$. The existence of a latent heat at melting $L_1 = T_{m1}\Delta S_1$ should be revealed by a plateau at $T_e = T_{m1}$ in the time evolution of the common electronic temperature T_e for some time during the heating pulse and during the subsequent relaxation. If P is the absorbed power, these plateaus would have widths

$$\Delta t_+ = L_1/[P - K_{e\text{-He}}(T_{m1} - T)], \quad (6)$$

$$\Delta t_- = L_1/K_{e\text{-He}}(T_{m1} - T), \quad (7)$$

respectively, upon heating and upon subsequent relaxation to the helium bath. $K_{e\text{-He}} = K_{1\text{-He}} + K_{2\text{-He}}$ is the total electron-helium thermal conductivity.

To realize the two-density configuration, the top electrode is divided into a small disk of radius $R_1 = 6.3$ mm and a ring $R_1 < r < R_2 = R$ kept respectively at potentials $-V_1$ and $-V_2$. Two concentric regions of density n_1 and n_2, respectively, on the same helium surface are obtained such that $n_2 - n_1 = (V_1 - V_2)/2\pi eD$. This relation, as well as the existence of two distinct triangular lattices at low temperature ($T \ll T_{m1} < T_{m2}$), has been checked at high densities by the detection of resonances near the ripplon frequencies corresponding to the reciprocal-lattice wave numbers for both crystals. At the low densities required for the present experiment, plasmon resonances are excited and detected as before. Most of the plasmon modes extend over the two regions and the frequencies become

$$\omega_{\nu,\mu} = (\omega_{p\nu,\mu}^2 + \alpha_{\nu,\mu}\omega_0^{(1)2} + \beta_{\nu,\mu}\omega_0^{(2)2})^{1/2}, \quad (8)$$

where $\omega_0^{(1)}$ and $\omega_0^{(2)}$ are the local modes of the low- and high-density regions; $\omega_{p\nu,\mu}$ is now a function of n_1 and n_2, while α and β are (calculable) weights reflecting the different wave amplitudes in the two portions.

The calorimeter is heated by a pulse on resonance with a mode whose amplitude is well localized over the ring. Its temperature is measured by the monitoring of a second mode well localized in this region whose frequency variation is sensitive to T_2 but not to T_1. To check that thermal equilibrium between calorimeter and sample is achieved, we have also measured T_1 by the frequency variation of a mode distributed over both regions and sensitive to both T_1 and T_2. The agreement for the temperature range near and below T_{m1} is rather good: $T_1 = T_2(1^{+0}_{-0.1})$. Also, when the helium temperature T is set below T_{m1} and the calorimeter is continuously heated, melting of the test sample is signaled by the vanishing of $\omega_0^{(1)}$ at a temperature $T_2 = T_{m1}$.

The time evolution of the electronic temperature is shown in Fig. 4: The electrons are heated from equilibrium with the helium substrate at $T = 135.5 \pm 0.3$ mK to a temperature $T_e(\max) = 160$ mK and subsequently allowed to relax. The melting temperature $T_{m1} = 149.5 \pm 1$ mK is thus crossed on both heating and cooling. There is no evidence of a plateau or of a change in slope at melting. We can set upper bounds

$$\Delta S < 0.2k_B; \quad [C(T_m^+) - C(T_m^-)]/C(T_m) < 0.09 \quad (9)$$

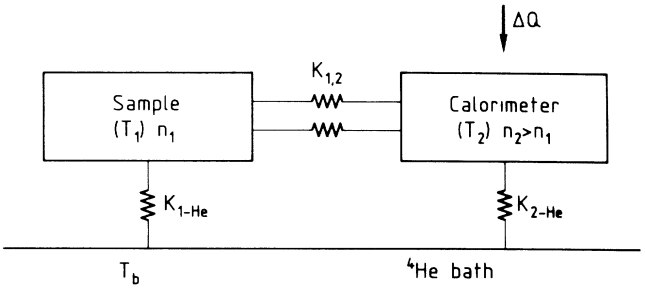

FIG. 3. Thermal schematic for experiments near melting. The calorimeter by which heating and temperature measurement are effected is well in the solid phase at melting of the sample. Thermal contact is assured by a common boundary (1D Kapitza resistance).

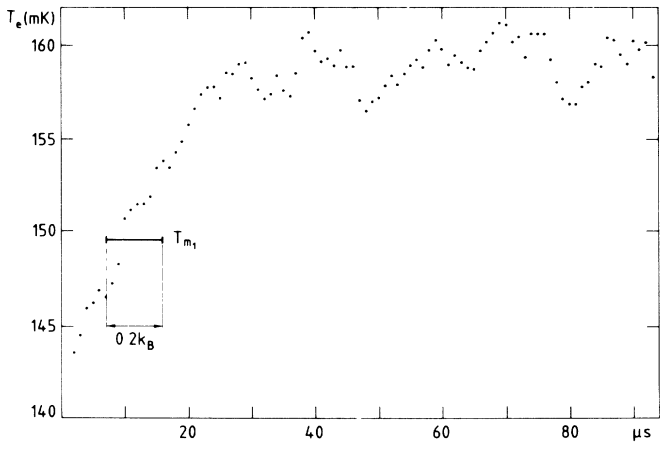

FIG. 4. Calorimeter (and sample) temperature during heating by a 6.1-pW pulse. A latent heat of $0.2k_BT_{m1}$ would give rise to a plateau whose width is shown by the bar at T_{m1}. The sample density is $n_1 = 0.44 \times 10^8$ cm^{-2} and the calorimeter density $n_2 = 1.0 \times 10^8$ cm^{-2}.

per electron, based on a minimum detectable plateau width of 8 μs estimated from the signal-to-noise ratio.

The difference between a first- and higher-order transition is in practice a quantitative question of setting a limit on the discontinuity of the entropy or its derivatives. We feel that to establish an upper limit which is lower than the entropy jumps ($\simeq 0.3 k_B$) usually found in numerical simulations,[4,12] is a reasonable criterion, and from there we conclude that melting of the 2D $1/r$ system is not first order.

Work is continuing to refine the results for $C(T)$ near T_m to an accuracy comparable with the low-temperature values and to extend the measurements further into the liquid phase to see if there is a peak associated with free dislocations.

We are pleased to acknowledge helpful discussions with D. Marty, J. Poitrenaud, V. Shikin, and especially G. Deville. Our thanks go also to C. Chaleil-Heyer and J. Verrier for invaluable technical help.

---

[a]Present address: Serin Physics Laboratory, Rutgers University, Piscataway, NJ 08854.

[1]J. M. Kosterlitz and D. J. Thouless, J. Phys. C **6**, 1181 (1973); D. R. Nelson and B. I. Halperin, Phys. Rev. B **19**, 2457 (1979); A. P. Young, Phys. Rev. B **19**, 1855 (1979).

[2]S. T. Chui, Phys. Rev. Lett. **48**, 933 (1982); Y. Saito, Phys. Rev. B **26**, 6239 (1982).

[3]T. V. Ramakrishnan, Phys. Rev. Lett. **48**, 541 (1982).

[4]J. Tobochnik and G. V. Chester, Phys. Rev. B **25**, 6778 (1982); S. W. Kock and F. F. Abraham, Phys. Rev. B **27**, 2964 (1983); A. F. Bakker, C. Bruin, and H. J. Hilhorst, Phys. Rev. Lett. **52**, 449 (1984), and references therein.

[5]For example: Continuous: P. A. Heiney, P. W. Stephens, R. J. Birgenau, P. M. Horn, and D. E. Moncton, Phys. Rev. B **28**, 6416 (1983); T. F. Rosenbaum, S. E. Naglar, P. M. Horn, and Roy Clarke, Phys. Rev. Lett. **50**, 1971 (1983); H. K. Kim, Q. A. Zhang, and M. H. W. Chang, Phys. Rev. Lett. **56**, 1579 (1986). First order: S. B. Hurlbut and J. G. Dash, Phys. Rev. Lett. **53**, 1931 (1984).

[6]C. C. Grimes and G. Adams, Phys. Rev. Lett. **42**, 795 (1979).

[7]G. Deville, A. Valdes, E. Y. Andrei, and F. I. B. Williams, Phys. Rev. Lett. **53**, 588 (1984); F. Gallet, G. Deville, A. Valdes, and F. I. B. Williams, Phys. Rev. Lett. **49**, 212 (1982).

[8]D. C. Glattli, E. Y. Andrei, G. Deville, J. Potrenaud, and F. I. B. Williams, Phys. Rev. Lett. **54**, 1710 (1985).

[9]D. S. Fisher, B. I. Halperin, and P. M. Platzman, Phys. Rev. Lett. **42**, 798 (1979).

[10]L. Bonsall and A. A. Maradudin, Phys. Rev. B **15**, 1959 (1977).

[11]D. S. Fisher, B. I. Halperin, and R. Morf, Phys. Rev. B **20**, 4692 (1979).

[12]R. C. Gann, S. Chakravarty, and G. V. Chester, Phys. Rev. B **20**, 326 (1979); R. H. Morf, Phys. Rev. Lett. **43**, 931 (1979); R. K. Kalia, P. Vashishta, and S. W. de Leeuw, Phys. Rev. B **23**, 2794 (1981).