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STRUCTURE OF THE ELECTRON GAS AT THE SURFACE OF LIQUID HELIUM

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Abstract.—A brief, tutorial survey is presented of the structure of the two-dimensional electron gas formed by electrons in image-potential induced surface states on liquid helium. Emphasis is placed on the change in structure as the electronic area density is varied and the system properties change from those of an electron gas to an electron liquid and finally to an electron crystal.

1. INTRODUCTION.—An electron in the vapor above a liquid helium surface is attracted toward the surface by the classical image force. However, within a few Angstroms of the surface the electron experiences a strong repulsive barrier to penetration into the helium. The repulsive barrier arises from the Pauli exclusion principle which requires that the wave function of an excess electron in the liquid He must be orthogonal to the wave functions of the electrons on the He atoms. The potential well formed by the combination of the image potential and the repulsive barrier supports a series of bound electron states which are similar to the s states of the hydrogen atom, but bound with four orders of magnitude less energy. At 1 K and below, such surface electrons are predominantly in the ground state within the image-potential well and are localized within ± 100 Å of the helium surface, but they are not localized in the plane of the surface and remain free to move parallel to the surface. A sheet of electrons on a liquid helium surface then forms a classical two-dimensional one-component Coulomb gas of variable density.

The purpose of this paper is to provide a brief tutorial review of what is known and what has been predicted concerning the structure of the two-dimensional electron gas on a helium surface. The emphasis will be on the variation in properties as the electron area density is increased at low temperatures and the behavior changes progressively from electron gas to liquid to solid.

The possibility that the potential well outside a liquid helium surface could support a series of bound electron states was independently realized by Sommer /1/ and by Shikin /3/. The experimental investigations reported measurements of the mobility /4/ and the trapping lifetime /5/. Subsequent experiments have included cyclotron resonance /6,7/ and a study of plasmon dispersion and damping /8/. These experiments have confirmed the existence of surface state electrons (SSE), demonstrated that the effective mass is very nearly the free electron mass, measured the binding energy and Stark tuning rate, and measured the electron mobility in different temperature regimes. At low temperatures (below 0.7 K) the SSE mobility parallel to the helium surface, which is limited by electron-rippon scattering, has a value near 10^7 cm²/Vs and decreases approximately as 1/T for increasing temperature, T. Above about 0.8 K the SSE mobility is limited by gas atom scattering and decreases nearly exponentially with increasing T.

The thermodynamic state of the 2D electron gas is determined by the ratio of the potential energy per electron to the kinetic energy. This dimensionless ratio, given by $\Gamma = \frac{n_s}{n_{th}}$, characterizes the degree to which the electronic motions are correlated. When $n_s$ is small, i.e. $\Gamma \lesssim 1$, the kinetic energy dominates and the SSE behave as a 2D classical electron gas. At higher densities and lower temperatures ($1 \lesssim \Gamma \lesssim 100$) the electronic motions become more correlated and liquid-like behavior is expected. At still higher densities and low temperatures a 2D electron crystal is expected to form (for $\Gamma \gtrsim 100$).
SSE obey Maxwell-Boltzmann statistics. This will be true as long as $kT \gg EF$ where the Fermi energy for a 2D system is given by $E_F = \pi k^2 n_p / m$.

For surface electrons on a liquid there exists an upper bound on the attainable electron areal density. Gorkov and Chernikova /10/ pointed out that the expression for the frequency of coupled plasmons and ripplons on a liquid surface contains competing terms that can cancel and cause the frequency to go to zero for a finite wavelength at a critical electron areal density. The critical density is determined by the liquid's surface tension and for $^4$He has the value $N_s = 2.2 \times 10^9$ cm$^{-2}$. At this areal density the Fermi temperature is only 18 mK, so the SSE on He obey Maxwell-Boltzmann statistics at all the temperatures studied thus far ($T > 0.3$ K). For the remainder of this paper we assume $kT/E_F \gg 1$.

In the remainder of this paper we present in succession brief sections on the 2D electron gas, liquid, and crystal. Each section contains first a mention of the relevant theoretical works, then a summary of the pertinent experiments - if any - that have been performed, and finally mention is made of some obvious experiments remaining to be done.

2. THE LOW DENSITY 2D ELECTRON GAS.- Several theoretical papers have appeared recently which have treated explicitly the classical 2D electron gas. As mentioned above we expect gas-like behavior for $\Gamma \leq 1$. This expectation is borne out in calculations of the pair correlation function by Totsuji /11/ and by Lado /12/. Totsuji performed numerical experiments by the Monte Carlo method using 81 particles within a unit cell in a periodic system. He found that for $\Gamma \leq 1$, the pair correlation function is a monotonically increasing function of separation. Oscillatory structure in the pair correlation function, indicating the onset of short range order, first appeared for $2.2 < \Gamma < 2.7$. Very similar results were obtained by Lado who solved numerically the hypernetted-chain integral equation. He also found gas-like behavior for small $\Gamma$, but oscillatory structure in the pair correlation function first appeared for $2.8 < \Gamma < 2.9$ which is a slightly higher value than found by Totsuji. Chalupa /13/ Totsuji /11,14/ and Fetter /15/ have calculated the equation of state for the classical 2D electron gas. They find that the leading corrections to the ideal gas equation of state are of order $\Gamma^2$.

The electrodynamic and thermodynamic properties the 2D classical electron gas have been calculated by Fetter /15/ and by Totsuji /14/. They both employed many-body techniques to calculate, in the random phase approximation, the screening of a static impurity, the spectrum and damping of plasma oscillations, and inelastic electron scattering. The calculated plasmon dispersion relation explicitly displays the effects of Landau damping. They obtained the equation of state and other thermodynamic functions by Debye-Hückel type expansions.

Few experiments have been performed in the regime where $\Gamma \leq 1$. Such experiments require small areal densities of electrons. For example: consider electrons on a liquid He surface at $T = 4$ K, then the condition $\Gamma \leq 1$ requires $n_s \leq 10^5$ cm$^{-2}$ which corresponds to an externally applied holding electric field of $E = 1$ V/cm. In a typical experimental cell which is 0.3 cm high, the applied potential would be only 0.03 V. Thus to manipulate such small surface charge densities requires care to avoid extraneous potential offsets which can arise from thermal EMFs and charge inadvertently deposited on the He film which coats all surfaces in the experimental cell. Measurements employing small surface charge densities, also, of course, yield relatively small signals.

For the reasons mentioned above, most experiments have been performed in the regime $10 \leq \Gamma \leq 100$. However, Bridges and McGill have performed a time-of-flight mobility measurement using small areal densities of electrons on He /16/. They employed pulses of electrons consisting of about 30 to 600 electrons per pulse which occupied an area of 0.2 cm$^2$ resulting in $n_s < 10^6$ cm$^{-2}$. Their lowest temperature was about 1.2 K yielding $\Gamma \leq 0.2$. At 1.2 K their electron mobility fell significantly below the mobility obtained by others at the same temperature but with densities in the electron liquid regime /4,6,17/ Platzman et al. /18/ have calculated the density and frequency dependence of the conductivity when ripplon scattering is dominant. They find that the electron mobility should be an increasing function of density due to screening of the ripplons by the electrons. However, due to a finite frequency approximation that they employed, their calculation is not directly applicable to the Bridges and McGill experiment. Further work is needed to establish if the observed mobility is an "electron gas" effect.
There are a number of interesting experiments remaining to be performed on the classical 2D electron gas. Observation of the effects of Landau damping on plasmon dispersion and damping would be of considerable interest. In such an experiment one would be of considerable interest. In such an experiment one would expect to see the predicted transition from the plasmon mode \( \omega_p = k^{1/2} \) to a sound-like collective mode at low areal densities /15,19/. Any experiment, such as light scattering, that could yield information on the structure factor would be of great interest. Fetter has pointed out that the 2D electron gas may be more easily studied by using Ne as a substrate instead of He /15/. By working with solid Ne near \( T = 10^0 \), much higher areal densities (\( n < 10^7 \)) could be used and still have \( \Gamma \leq 1 \).

3. THE CLASSICAL 2D ELECTRON LIQUID.- In the electron liquid regime \( 1 \leq \Gamma \leq 100 \), no small expansion parameter exists so calculations are difficult. Recently, however, there have appeared three computer calculations on the structure of the 2D electron liquid: Totsuji's Monte Carlo calculation /11/ which treated the parameter range \( 0.16 \leq \Gamma \leq 50 \), Lado's hypernetted chain calculation /12/ which spanned \( 0.1 \leq \Gamma \leq 100 \), and the molecular dynamics calculation by Hockney and Brown /20/ which spanned the range \( 33 \leq \Gamma \leq 1850 \). As mentioned above, the pair correlation function starts to exhibit oscillatory structure as \( \Gamma \) exceeds the value 2.7.

As \( \Gamma \) increases, the short range order increases and the amplitude of the oscillatory structure in the pair correlation function increases. The gradual growth in short range order with increasing \( \Gamma \) is illustrated by the schematic pair correlation functions shown in figure 1. The structure factor displays a similar growth in oscillatory structure with increasing \( \Gamma \). The physical picture suggested by the calculations is one in which it is becoming increasingly probable that a given electron will be surrounded by six nearest neighbors each lying close to the sites of a triangular lattice, but without having any long-range correlation in positions. At large values of \( \Gamma \) (near \( \Gamma = 100 \)) one can visualize each electron as oscillating rapidly in a potential well due to its neighbors as it wanders slowly from one configuration to another.

Although most of the experiments on SSE on liquid He have been performed in the electron liquid regime, only one experiment, that of Zipfel, Brown and Grimes (ZBG) has yielded direct information pertaining to the short range order of the electron liquid /21/. That experiment was an extension of a study mentioned in the introduction regarding the spectra of transitions between bound electronic states in the image potential-well /8/. The earlier experiments measured the splittings between the ground state and the first two excited states by observing mm wave absorption as the splittings were Stark-tuned into resonance with the frequency of the applied mm radiation. By applying a magnetic field \( B \parallel \) parallel to the liquid He surface, ZBG obtained from the spectroscopy experiment a time, \( \tau \), that is expected to be closely related to the electron velocity auto-correlation time. Applying \( B \parallel \) couples the spectroscopic transitions to the component of the electronic motion parallel to the He surface but perpendicular to the magnetic field. Physically, this coupling is due to the Lorentz force which is equivalent to an electric field normal to the He surface and thus acts as a perturbation to the Stark tuning field. Since this perturbation is proportional to a component of the electron's velocity, we expect that at low SSE areal densities application of \( B \parallel \) will simply broaden the spectroscopic line-shape into a Gaussian whose width is a measure of the temperature of the electron gas. However, at high areal densities the electron-electron scattering becomes greater and the contribution...
to the linewidth due to $B_{pl}$ is appreciably reduced by motional averaging. In the motionally narrowed regime the linewidth contains a contribution that is proportional to $\tau_C$. Values of $\tau_C$ at different areal densities were deduced from the observed linewidths, and were found to decrease from $9.9 \times 10^{-11}$ s to $1.4 \times 10^{-11}$ s as $\Gamma$ increased from 9 to 36. ZBG made the physically plausible assertion that $\tau_C$ is the electron velocity auto-correlation time. They also pointed out that the observed values of $\tau_C$ are close to the periods expected for harmonic oscillation of an electron about a lattice site in a triangular electron lattice of the same areal density.

Itoh et al. derived for the 2D electron liquid an expression that related the velocity auto-correlation time to the radial distribution function. Using the pair correlation functions from the computer calculations of Hockney and Brown and of Totsuji they then calculated the velocity auto-correlation times at several values of $\Gamma$. The velocity auto-correlation times thus obtained agreed with the values of $\tau_C$ measured by ZBG within the experimental error of $\pm 10\%$. Itoh et al. also used a harmonic lattice model to construct an approximate radial distribution function which was then employed to calculate auto-correlation times. The auto-correlation times thus obtained were about 30% larger than the measured values of $\tau_C$. They conclude from the close correspondence of these numbers that the short range order in the electron liquid even at $\Gamma = 36$ is much like that in the electron crystal.

Platzman and Halperin (PH) have examined in detail the quantities that can be measured in the ZBG type of experiment. They find that the interpretation of the experiment is complicated in all but a few limiting cases. Omitting ripplon and vapor atom scattering, they find the interpretation is simple in the low density limit $\Gamma \ll 1$. In this case the approach taken by ZBG is borne out: the broadening of the lineshape by the applied field is partially removed by motional narrowing leading to a linewidth proportional to $\tau_{ac}$, the velocity auto-correlation time.

At higher densities in the electron liquid regime, the velocity auto-correlation function is expected to develop oscillatory structure just as the pair correlation function does. (Hansen, McDonald and Pollock have calculated velocity auto-correlation functions for the three dimensional electron liquid and find that as $\Gamma$ increases the oscillatory contribution from the plasmon increases). The PH analysis then indicates that the absorption lineshape should no longer be simple, but is related to the Fourier transform of the velocity auto-correlation function and therefore contains structure. The interpretation of experimental lineshapes should then be more complicated than the procedure used by ZBG. However, in view of Itoh et al.'s finding of close agreement between the ZBG $\tau_C$ and the $\tau_{ac}$ calculated from the pair correlation functions, it appears that $\tau_C$ is actually a measure of the velocity auto-correlation time. The extra structure in the lineshape predicted by PH was not seen in the experimental traces.

Platzman and Halperin emphasize that the ZBG type of experiment, if performed at lower temperatures and in a larger range of magnetic fields, could yield information on the dynamical properties of the electron liquid or electron crystal. The lineshape should contain information on self-diffusion in the liquid and on transverse modes, defect motion and zone boundary phonons in the solid.

The effects of electron-ripplon and electron-He atom scattering on spectroscopic linewidths both with and without an applied magnetic field have been analyzed by Ando. His results, which are applicable in the low-density, $\Gamma \ll 1$, regime emphasize that the linewidth contains contributions from broadening of both the ground state and the excited state. His calculations should prove very useful when the spectroscopy experiments are extended to lower temperatures and lower electron densities.

Many other experiments performed in the liquid regime have not yet yielded information on the structure of the liquid. For example, cyclotron resonance and mobility measurements have yielded only "one electron" properties of the system. Experimental studies of the long wavelength plasmon dispersion, although they probe a collective mode of the system, are not sensitive to the short-range structure. In fact, the long wavelength plasmon dispersion relation remains unchanged throughout the liquid and crystal regimes. Of course, if the plasmon studies could be extended to short wavelengths near the Brillouin zone boundary, then the plasmon dispersion becomes sensitive to the short range structure.
charge densities. At high charge densities ($\Gamma \approx 36$) the spectroscopic line develops side bands that move out with increasing $n_{pl}$ /26/. These sidebands may be due to "zone boundary" plasmons in the highly correlated liquid /27/.

Clearly, there are many interesting experiments remaining to be performed on the electron liquid. The spectroscopy-in-a-magnetic-field (ZBG) type of experiment is very promising and warrants additional work. Again, any experiment that could measure the structure factor would be very useful.

4. THE HIGH DENSITY 2D COULOMB CRYSTAL.- At high electron areal densities, $\Gamma \lesssim 100$, the Coulomb potential energy predominates and the state of lowest energy becomes the triangular electron lattice. This 2D analogue of Wigner crystallization was first discussed by Grandall and Williams /28/ while the phase diagram was first calculated by Platzman and Fukuyama /29/. The Hockney and Brown (HB) molecular dynamics calculation places the melting transition at $\Gamma = 95$ /20/. The phase diagram based on that value is sketched in figure 2. One of the most interesting result of the HB computer experiment is the discovery of the lambda-type anomaly in the electronic specific heat at the liquid-solid phase transition. They find that all other calculated properties (internal energy, pair correlation function, structure factor) vary smoothly through the phase transition, so it appears to be a continuous liquid-solid phase transition.

To date, no firm laboratory experimental evidence for the existence of the 2D electron crystal has appeared. Nevertheless, the concept of electron crystallization has stimulated many persons to write theoretical papers. A partial listing follows:

The phonon spectra of triangular /29,30/ and square /30-33/ electron lattices have been calculated, and the square lattice has been shown to be dynamically unstable /30,33/. The longitudinal phonon mode of the electron crystal is just the 2D plasmon $\omega^2 = 2\gamma_{pl} e^2/k/m$ which is dispersive, whereas the transverse mode $\omega = ck$ is sound-like at long wavelengths. A magnetic field applied normal to the He surface couples the two phonon modes /30,32,34,35/.

In a strong electric field $E_{||}$, a dimple can form in the He surface under each electron and form a harmonic oscillator potential well. The dispersion relations for the coupled harmonic oscillator and phonon modes have been derived by Shikin /36/.

Shikin has proposed a novel experiment to detect the presence of the electron crystal on liquid He /37/. He suggested using the periodic electron lattice as a transducer to excite standing capillary waves on the He surface. Driving the electron crystal with an rf field applied normal to the surface should excite resonances when the frequency and the low index reciprocal lattice vectors $q_{\parallel j}$ of the electron lattice satisfy the capillary wave dispersion relation $\omega^2 = (\alpha/\rho) q_{\parallel j}^4$ where $\alpha$ is the surface tension coefficient of He and $\rho$ is the density. This proposed experiment was analyzed in detail by Monarkha and Shikin /38/. If observed, the resonances would provide clear evidence for the existence of the electron crystal.

5. CONCLUSION.- It is clear that much more theoretical work has been done on the 2D electron system than experimental work. There is an evident need for experimental tests of the theoretical models and predictions.

If the reader has been intrigued by this simple, but many faceted system, and would like to read further, an uncritical review with a fairly complete list of references appeared recently /39/. Earlier reviews were written by Shikin and Monarkha /40/ Crandall /41/ and Cole /42/.
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