Observation of Cyclotron Resonance in Surface-Bound Electrons on Liquid Helium

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We have observed cyclotron resonance from electrons in two-dimensional states outside liquid helium. Their resonant frequency depends only on the perpendicular component of the applied magnetic field, and their observed mobility is that expected for electrons scattered parallel to the liquid surface in only two dimensions.

In a recent Letter, Ostermeier and Schwarz¹ have presented data questioning the existence of image-potential-induced surface states for electrons outside of liquid helium. These states had been predicted for dielectrics generally by Cole and Cohen.² They are caused by the potential well formed by the attractive image potential outside a dielectric surface, and the repulsive barrier to electron penetration at the surface. For liquid helium, they are calculated to have a binding energy of 0.68 meV and a spatial extent outside the liquid surface of approximately 100 Å. Observation of these states was reported by Sommer and Tanner³ and Williams, Crandall, and Willis (WCW).⁴ Sommer and Tanner measured the mobility of electrons trapped in surface states by observing the phase shift between drive and response voltages on electrodes just under the charged liquid surface. They observed a mobility a factor of 4 or 5 smaller than that observed for electrons in gaseous helium at the same temperature. WCW observed the lifetimes of electrons on the surface by removing their holding field for a few microseconds and then measuring the charge remaining on the surface. They observed a temperature-independent lifetime for electrons outside the surface of approximately 100 μ sec. Ostermeier and Schwarz, with a single apparatus, attempted to repeat both of the above measurements. They used a time-of-flight method to measure electron mobilities parallel to the surface. By merely observing the reduction in the charge reaching their collector after they had removed their holding field for a short time, they could also repeat the lifetime measurements of WCW. They found no evidence for a different mobility for the surface-state electrons from those in the gas, and set an upper limit of a few microseconds on the lifetimes of such surface states. In view of this experimental disagreement we wish to report preliminary measurements of cyclotron resonance (CR) from electrons trapped on a liquid-helium surface. To summarize our results, we demonstrate the existence of electrons in two-dimensional (2D) states and observe mobilities comparable to those measured by Sommer and Tanner. We also calculate a 2D mobility and find reasonable agreement with our measurements and those of Sommer and Tanner.

Our experimental apparatus consists of a microwave spectrometer with its cavity mounted in a can partially filled with liquid helium and which is immersed in a pumped helium bath. The cavity is a right circular cylinder 1.2 cm in diameter and 0.75 cm in height split at the midplane where a Mylar sheet full of small holes is inserted to damp sloshing of the liquid helium. The cavity resonates at 23.5 GHz in the TE_{111} mode with the microwave electric field parallel to the liquid surface which is adjusted to lie in the upper half. By applying a voltage between the halves of the cavity, electrons can either be attracted to or repelled from the liquid surface. A sharp point can be inserted into either the side of the upper half of the cavity or the wave guide just above the cavity and is used to initiate a glow discharge which serves as an electron source above the liquid surface. A magnetic field up to 17 kOe produced by a superconducting Helmholtz pair can be applied vertically or tilted up to 28° to the vertical.

After trapping electrons on the surface by manipulating the holding electric field (this procedure will be discussed below), the glow is turned off and not turned on again until we wish to recharge the surface. A typical example of the power absorbed in the cavity as a function of magnetic field is presented in Fig. 1 for two different orientations of the field with a holding voltage of 250 V. (The lines in the figure are power broadened.) The amplitude of the signal⁵ corresponds to a charge density of 3×10^8 to 10^9 electrons/cm². As can be seen, the peak in the resonance curve is shifted toward higher fields when the field is rotated away from the vertical. Figure 2 is a plot of the ratios of the fields at shifted peaks to



FIG. 1. Typical examples of cyclotron-resonance absorption lines for surface-state electrons on liquid helium in tilted and untilted fields.

the field at the unshifted peak versus $\sec \theta$. The straight line is the expected behavior for an electron confined to move only in the plane of the surface.⁶ We feel that this is compelling evidence for the two-dimensional character of these states.

The mobilities of these electrons at various gas densities were obtained from our observed unbroadened⁷ CR linewidths⁸ and are presented in Fig. 3 along with the mobilities measured by Sommer and Tanner.⁹ Also presented in Fig. 3 are two theoretical mobilities, one due to the scattering of electrons from helium atoms in the gas, the other due to the same scattering process, but with the electrons confined to move along the liquid surface without change of their perpendicular eigenstate.¹⁰ The expressions for the mobilities in the two cases are

 $\mu_{2D} = (e/\pi\hbar\sigma n) [\int_0^\infty \varphi^4(z) dz]^{-1},$ $\mu_{3D} = \frac{4}{3} e/n\sigma (2\pi mkT)^{1/2},$

where *n* is the density of helium atoms, σ the cross section, ¹¹ $\varphi(z)$ the perpendicular wave function, and the other symbols have their usual meanings. To obtain numerical results the wave function $\varphi(z)$ was taken to be $2a^{-3/2}z \exp(-z/a)$, where a = 78 Å. This corresponds to assuming an infinite barrier at the liquid surface with an image potential of $-e^2[(\epsilon-1)/4(\epsilon+1)]z^{-1}$ outside the helium. We emphasize that there are no free parameters in this calculation. We feel the agreement between our measurements and this simple theory to be quite good and another strong piece of evidence for the 2D character of these states.

We now describe the procedure used to charge the liquid surface. Initially, we expected to be able to charge the surface merely by applying a positive voltage to the bottom half of the cavity



FIG. 2. Relative peak shift versus $\sec\theta$, where θ is the angle of tilt of the magnetic field. The error bars are $\pm 0.5^{\circ}$ horizontally and represent maximum uncertainty vertically. Data points are from several runs and temperatures ranging from 1.55 to 1.2°K. The straight line is the expected behavior for an electron in a purely 2D state.

while the glow was on. However, by monitoring the microwave power absorbed in the cavity, we found that in the steady state when the glow was on it was not possible to keep the surface charged. If a positive voltage was suddenly (in a fraction of a second) applied to the bottom half of the cavity, electrons would appear on the liquid surface and then slowly decay away with a time constant roughly proportional to the inverse of the discharge current.¹² If the discharge was turned off while electrons were still on the surface, the decay stopped and the electrons could be held for periods of hours with no loss. At present, we have no explanation for this effect. It may be related to the presence of positive ions, recombination photons, or just heat from the glow, or it may be due to charging of the helium films present on the walls of the cavity. We can observe this latter effect by first reducing the holding field (after the glow is off) and then increasing it again. This causes the number of electrons on the surface first to decrease and then, when the field is raised again, to increase to or near its original value. The released electrons cannot be in the gas above the surface as they would appear as an unshifted cyclotron line in a tilted field



FIG. 3. Electron mobility versus helium gas density. Circles, this work; crosses, Sommer and Tanner (Ref. 3); solid line, 2D theory; dashed line, 3D theory.

which we do not observe. Presumably, therefore, they are on the films covering the walls of the cavity where they cannot be influenced by the microwave electric fields.

These obviously complex charging effects are one possible explanation for the disagreements between the previously reported experiments. If helium films in unexpected parts of an apparatus can be charged with mobile electrons, then it is difficult if not impossible to predict the actual electric fields present for a given set of externally applied voltages.

In our own experiment this uncertainty leads us to point out that we nay be observing electricfield-induced surface states rather than those induced by the image potential. Several observations, however, cause us to believe this is not the case. First, since the number of electrons decreases as we decrease the holding potential, it seems reasonable to assume that the surface is fully charged and thus cancels the electric field above it. In this case there would be no electric field far from the surface and, although the exact potential near the surface is difficult to calculate, a rough estimate indicates the binding energy is charged by less than 20% for the holding fields used in our experiment. Thus, the image potential dominates in determining the properties of the state. This is supported by the fact that we observe no change in the position or shape of the shifted CR line (aside from a decrease in intensity) when we reduce the holding field by a factor of 10. Finally, there is the numerical agreement between our mobility measurements and the 2D scattering calculation with no adjustable parameters.¹³ We feel that the above arguments, taken together, are reasonably persuasive, and therefore feel that the best way to explain our data is in terms of image-potential-induced surface states.

In summary, we feel we have demonstrated conclusively the existence of electrons in 2D states outside of liquid helium. Although it is not possible to conclude definitely that these are imagepotential-induced states rather than electricfield-induced states, we believe the former to be the most reasonable assumption. In any case, many of the most physically interesting properties of these electrons, such as the fact that here one has a 2D electron system with a density variable over many orders of magnitude, or that they may be used as a probe of the liquid-helium surface, independent of the nature of the binding potential.

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¹R. M. Ostermeier and K. W. Schwarz, Phys. Rev. Lett. 29, 25 (1972).

²M. W. Cole and M. H. Cohen, Phys. Rev. Lett. <u>23</u>, 1238 (1969). See also M. W. Cole, Phys. Rev. B <u>2</u>, 4239 (1970).

³W. T. Sommer and D. J. Tanner, Phys. Rev. Lett. <u>27</u>, 1345 (1971).

⁴R. Williams, R. S. Crandall, and A. H. Willis, Phys. Rev. Lett. <u>26</u>, 7 (1971).

⁵Since the radial distribution of charge is unknown, its effects on the cavity can only be roughly estimated by using a uniform charge distribution.

⁶We should mention at this point that we have observed CR in electrons outside helium films on a dielectric substrate where the field could be rotated to much larger angles. In this case, the shift in peak position is also proportional to $\sec\theta$ out to the highest fields we can apply, corresponding to an angle of 70°.

⁷Microwave heating of the electrons still caused slight power broadening of the resonance line even at the lowest power input (~ 10^{-8} W) consistent with reasonable signal-to-noise ratio. The unbroadened linewidths were obtained by extrapolating the observed power dependence to zero power input. This never caused a change of more than 20% from the last measured value. The extrapolation was assumed to double the measured uncertainties in linewidth.

⁸If the collision frequency ν is not independent of velocity, converting cyclotron-resonance linewidths to mobilities requires numerical integration [see F. Fehsenfeld, J. Chem. Phys. <u>39</u>, 1653 (1963)]. In 2D, ν is independent of v; in 3D $\nu = n\sigma v$. We find $\mu_{2D} = e\tau/m$ and $\mu_{3D} = 0.9e\tau/m$, where $\tau = 2Q/\omega$, ω being the experimental frequency, and Q has its usual meaning. The 2D relation was used to convert our data.

⁹W. T. Sommer (private communication) estimates that, in addition to 25% random error, there may have been systematic errors of comparable magnitude in his experiment. ¹⁰This calculation has been done previously by Cole (Ref. 2); however, we find our μ_{2D} to be a factor of 4 lower than his, using the same wave functions and electron-helium interaction (chosen to give the measured value of σ). See also R. S. Crandall, Phys. Lett. <u>37A</u>, 389 (1971).

 $^{11}\sigma = 4.9 \times 10^{-16} \text{ cm}^2$, B. Bederson and L. J. Kieffer, Rev. Mod. Phys. 43, 601 (1971).

¹²The same behavior was observed whether the discharge point was inside the cavity or in the wave guide although the decay rates were slower with the point in the wave guide. When the point was inside the cavity, the time constant was a few seconds at a discharge current of a few microamperes.

¹³Since μ_{2D} depends on $\int \varphi^4 dz$, it is sensitive only to the spatial extent of φ . A confining electric field of 50 V/cm would give approximately the same value for the integral as does the image potential.

Temperature Dependence of the Electrical Resistance for Isobutyric Acid–Water near the Critical Point

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The electrical resistance of two sealed samples of critical mixtures of isobutyric acidwater have been measured in the reduced temperature ranges $[\epsilon = T - T_c)/T_c] 6.7 \times 10^{-7} < \epsilon < 3.2 \times 10^{-2}$ and $3.3 \times 10^{-7} < \epsilon < 6.6 \times 10^{-2}$. Data analyses indicate that the temperature derivative of the resistance is strongly divergent as $\epsilon \rightarrow 0$.

Friedländer,¹ in his pioneering work, recognized a possible connection between the electrical resistance and the anomalously high shear viscosity for the electrolyte isobutyric acid-water in the critical region. He concluded qualitatively that the resistance was much less affected by critical behavior than was the viscosity. Recently, anomalies in the temperature derivative of the electrical resistance have been observed for ferromagnets near their Curie temperatures,^{2,3} antiferromagnets near their Néel points,⁴ and β brass near the order-disorder transition.⁵ In these materials the charge-carrying species are electrons which are scattered by local magnetic fluctuations that become very large near the second-order phase transition. de Gennes and Friedel⁶ and Fisher and Langer⁷ have presented theories to describe this phenomenon. The Fisher-Langer result that $\partial \rho / \partial T \sim (T - T_c)^{-\alpha}$, where α is the exponent describing the weak specificheat divergence, seems to be verified for the ferromagnet nickel.² In isobutyric acid-water the charge-carrying species are ions of opposite

sign, and the fluctuations near the critical mixing temperature are in the concentration or density. So far there are no theoretical predictions concerning the temperature dependence of the electrical resistance or its temperature derivative for a binary liquid mixture in the neighborhood of the critical point.

In this Letter we report on an experimental investigation of the electrical resistance and its temperature derivative for critical mixtures of isobutyric acid-water as the critical temperature is approached from the one phase (hightemperature) region.

The isobutyric acid and water used in this study were the same as those described in our earlier report⁸ on the viscosity of this system. The critical composition was 38.0% isobutyric acid by weight, and the critical temperatures were 26.238and 26.233°C for the two different samples used.⁹

The conductance cells were of a modified Jones-Bollinger¹⁰ design. The resistances of the cells were measured with a Jones¹¹ ac bridge utilizing a lock-in amplifier as a signal generator and