

Reprinted from: **LOW TEMPERATURE PHYSICS-LT 13, VOL. 1**

Edited by K. D. Timmerhaus, W. J. O'Sullivan and E. F. Hammel  
 Book available from: Plenum Publishing Corporation  
 227 West 17th Street, New York, New York 10011

## **Measurement of Ionic Mobilities in Liquid <sup>3</sup>He by a Space Charge Method\***

**P. V. E. McClintock**

*Department of Physics, University of Lancaster  
 Lancaster, England*

### **Introduction**

The behavior to be expected of a charged impurity moving in a Fermi fluid was first discussed in detail by Abe and Aizu<sup>1</sup> and independently by Clark.<sup>2</sup> They concluded that, although mobility should be inversely proportional to viscosity at high temperatures (classical limit), a  $T^{-2}$  law should be obeyed in the low-temperature (degenerate) limit. Davis and Dagonnier have discussed the situation at intermediate temperatures in terms of quantum mechanical Brownian motion.<sup>3</sup> These predictions were not borne out by experiment.<sup>4,5</sup> In a more recent theoretical investigation Josephson and Lekner<sup>6</sup> show that, although the  $T^{-2}$  law may still be expected at sufficiently low temperatures, some form of weaker dependence should be observed at higher temperatures. Since there seems to be little consensus as to the form of this weaker dependence or as to the characteristic temperature at which one regime might give way to the other, it is important that accurate experimental mobility data should be obtained. For negative ions such data are now available down to 17 m°K.<sup>5</sup> However, for positive ions data below 1°K,<sup>4</sup> measured by a time-of-flight method, suffer from severe inconsistencies and hysteresis effects, apparently experimentally based. In this paper we describe mobility measurements down to 0.25°K by a completely different technique: space-charge-limited emission of ions from a sharp metal point. This method appears to be more accurate than earlier space charge techniques<sup>7</sup> and avoids the complication of thermal gradients due to heating at a radioactive source.

### **Field Emission and Field Ionization in Liquid <sup>3</sup>He**

The characteristics of field emission and field ionization in liquid <sup>3</sup>He have been studied in detail.<sup>8,9</sup> In both cases there is a space-charge-limited regime for which the emission is well described by<sup>10</sup>

$$V = V_0 + 98(R/\alpha K\mu)^{1/2} i^{1/2} \quad (1)$$

where  $V$  is the potential in V applied to the tip,  $V_0$  is a constant,  $R$  is the anode radius

\* Work supported by the Science Research Council under contract BSR9251.

in cm,  $\alpha\pi$  is the emission cone angle in steradians,  $K$  is the dielectric constant,  $\mu$  is the mobility in  $\text{cm}^2 \text{V}^{-1} \text{sec}^{-1}$ , and  $i$  is the current in nA. In using Eq. (1) to determine mobilities, the largest uncertainty lies in  $\alpha$ . For emission in vacuum  $\alpha \approx 0.6$ , but in liquid helium we have found  $2.2 < \alpha < 3.2$ , the exact value depending on the individual tip. Since mobility *changes* smaller than 1% can be resolved, it is most useful to determine the temperature dependence of the mobility by our space charge method and then scale the data to agree with the absolute value of  $\mu$  at some particular temperature, obtained by another technique.

We have found that emission characteristics can be strongly influenced by the build up of thermal gradients in the liquid.<sup>9</sup> To avoid this effect, all data reported here were taken by applying potential to the tip only just long enough for a chart recorder to record the current, i.e., about 1.5 sec. A simple calculation shows that during this period the *average* temperature of the liquid rises by typically 30m°K. However, the maximum temperature rise probably occurs near the tip where the electric field is largest, and the change in temperature near the anode in the space charge region which controls the emission is presumably much smaller.

Details of the apparatus and experimental technique are discussed elsewhere.<sup>9</sup> The tungsten tips were prepared by electrolytic etching in NaOH solution and were smoothed *in situ* by field evaporation. Their radii of curvature were determined from the vacuum field-emission characteristics. Temperature measurement was by a 470- $\Omega$  Speer carbon resistor immersed in the sample within the anode assembly. It was calibrated against  $^3\text{He}$  vapor pressure down to 0.45°K and the calibration was then extrapolated to 0.25°K. Care was taken to keep the sample pressure constant since we have found that  $\alpha$  is weakly pressure dependent at low pressures.<sup>9</sup>

## Experimental Results

Current-voltage characteristics for both positive and negative emission are presented as  $i^{1/2}-V$  plots in Fig. 1 for three different temperatures. Good straight lines are obtained in agreement with Eq. (1), and the intercepts are  $V_0 = 410 \pm 20 \text{V}$  and  $490 \pm 30 \text{V}$  for negative and positive ions, respectively. Although we can, in

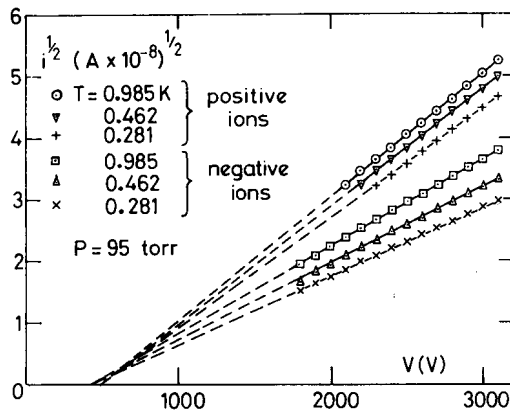


Fig. 1. Positive and negative ion emission (currents)<sup>1/2</sup> as a function of tip potential.

principle, determine  $\mu(T)$  by measuring the gradient at a number of different temperatures, this is not a convenient method in practice because of the large number of  $i(V)$  readings necessary to determine  $\mu$  at a single temperature.

If we rewrite Eq. (1) in the form

$$\mu(T) = [98^2 R / \alpha K (V - V_0)^2] i(T) \quad (2)$$

we see that at constant tip potential, changes in mobility are directly proportional to changes in emission current. It is convenient to normalize the current by dividing it by  $(V - V_0)^2$ , so that data taken at different tip potentials ought to fall on the same line. In Figs. 2 and 3 we plot the normalized negative and positive emission currents, respectively, against temperature for three different tip potentials in each case. Almost all the points fall within less than  $\pm 1\%$  of a "best fit" curve drawn through the data. Scaling our values of  $i/(V - V_0)^2$  to agree with earlier mobility measurements<sup>4</sup> at 1.0°K, we obtain the mobilities indicated by the right-hand scales of Figs. 2 and 3. The error in  $\mu$  relative to its value at 1.0°K is therefore indicated by the scatter of the data points and the systematic error in its absolute magnitude will be the  $\pm 10\%$  quoted by Anderson *et al.*<sup>4</sup>

## Discussion

From the linearity of the results shown in Fig. 1 we deduce that for both positive and negative ions the mobility is independent of electric field within our experimental range, i.e., up to several  $\text{kV cm}^{-1}$ , which is, as far as we are aware, the highest field for which the ionic mobility in liquid  $^3\text{He}$  has been measured. Careful scrutiny of the data suggests that the change in mobility resulting from a 50% change in average electric field is certainly less than 1%. Therefore the fact that the field in which  $\mu(T)$

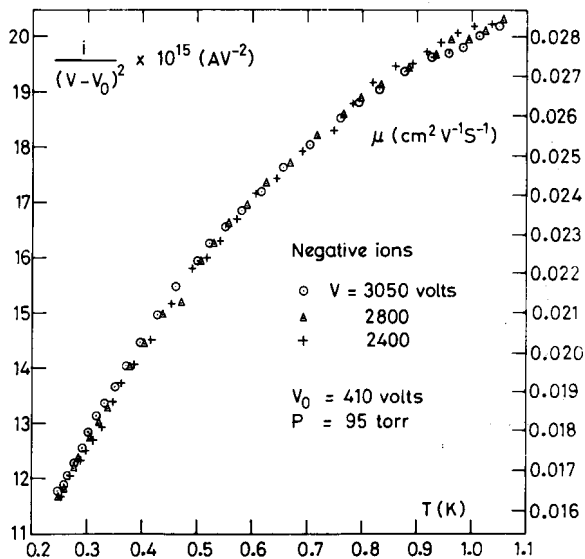


Fig. 2. Temperature dependence of the (normalized) negative ion emission current.

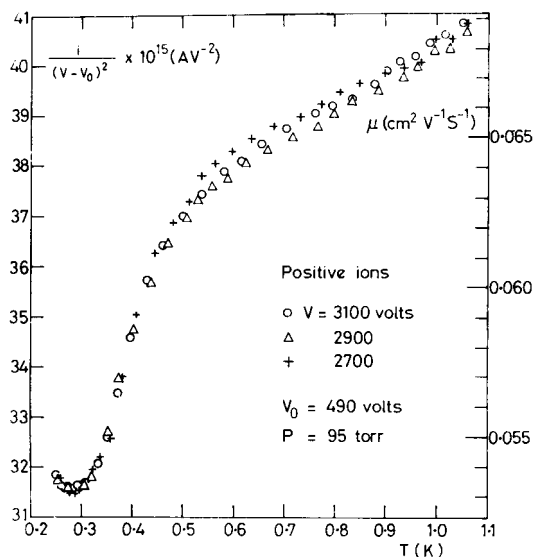


Fig. 3. Temperature dependence of the (normalized) positive ion emission current.

is measured is large and nonuniform has little effect on the reliability of our measurements.

The variation of  $\mu_-$  with temperature (Fig. 2) agrees well with the earlier results of Anderson *et al.*<sup>4</sup> in the region of overlap, but the scatter of our data points appears to be smaller. The temperature dependence of  $\mu_+$  (Fig. 3) is certainly not inconsistent with the earlier results,<sup>4</sup> but there is such a large scatter in the latter (about  $\pm 10\%$ ) that a meaningful comparison is difficult.

None of the existing theories predicts correctly the temperature dependence of  $\mu_+$  or  $\mu_-$  in this temperature range. Only Walden's rule gives the correct sign for the temperature dependence. Between 1.0 and 2.0°K we find  $\mu_-$  inversely proportional to viscosity within experimental error, using the smoothed viscosity data of Betts *et al.*<sup>11</sup> Below 1.3°K for  $\mu_+$  and 1.0°K for  $\mu_-$  the observed temperature dependence is much weaker than that implied by Walden's rule.

The minimum in  $\mu_+$  near 0.3°K was unexpected and is very interesting.\* A qualitative explanation can be given in terms of the "heliumberg" model of the positive ion.<sup>13</sup> On this model the minimum in the melting curve near 0.3°K<sup>14</sup> would imply a maximum in the positive ion radius at the same temperature and therefore, neglecting other effects, a minimum in mobility. However, it is hard on this basis to understand the shape of the  $\mu_+(T)$  curve. In particular, the maximum value of  $d\mu_+/dT$  occurs near 0.4°K, at which temperature the melting pressure is only 0.5% above its minimum value and is changing very slowly.

Our present mobility-measuring technique avoids complications arising from the heat continuously generated by a conventional radioactive source. In contrast, field emission/ionization sources generate no heat whatever except when actually

\* Kuchnir<sup>12</sup> reports recent observation of this minimum with an improved time-of-flight technique.

emitting current, and they would therefore seem particularly suitable for use in future experiments at ultralow temperatures. To extend our present investigation to lower temperatures, it is clear that much shorter current pulses will be required. Preliminary experiments in He I are encouraging: we find that pulse durations of 1 msec should be quite sufficient to carry out the measurements.

## Conclusion

Between 0.25 and 2.0°K the mobilities of both positive and negative ions in liquid  $^3\text{He}$  at low pressure are independent of electric field up to several  $\text{kV cm}^{-1}$ . We have measured the temperature dependence of the mobilities with greater resolution than that of previously published work. For negative ions our results are in excellent agreement with the earlier work. For positive ions they are not in disagreement but, with our superior resolution, we have been able to observe a definite minimum in  $\mu_+$  at around 0.3°K. No existing theory gives a satisfactory account of the mobility in this temperature range. In view of the great simplicity and good resolution of our space charge technique it seems highly desirable to extend the measurements down into the fully degenerate temperature regime, and we propose to do so at the earliest opportunity.

## References

1. R. Abe and K. Aizu, *Phys. Rev.* **123**, 10 (1961).
2. R. C. Clark, *Proc. Phys. Soc.* **82**, 785 (1963).
3. H. T. Davis and R. Dagonnier, *J. Chem. Phys.* **44**, 4030 (1966).
4. A. C. Anderson, M. Kuchnir, and J. C. Wheatley, *Phys. Rev.* **168**, 261 (1968).
5. M. Kuchnir, P. R. Roach, and J. B. Ketterson, *Phys. Rev.* **A2**, 262 (1970).
6. B. D. Josephson and J. Lekner, *Phys. Rev. Lett.* **23**, 111 (1969).
7. P. de Magistris, I. Modena, and F. Scaramuzzi, in *Proc. 9th Intern. Conf. on Low Temp. Phys. 1964*. Plenum Press, New York (1965), p. 349.
8. P. V. E. McClintock, *Phys. Lett.* **35A**, 211 (1971).
9. P. V. E. McClintock, *J. Low Temp. Phys.* **11**, 15 (1973).
10. B. Halpern and R. Gomer, *J. Chem. Phys.* **51**, 1031 (1969).
11. D. S. Betts, B. E. Keen, and J. Wilks, *Proc. Roy. Soc. A* **289**, 34 (1966).
12. M. Kuchnir, Private communication; M. Kuchnir, J. B. Ketterson, and P. R. Roach, this volume.
13. K. R. Atkins, *Phys. Rev.* **116**, 1339 (1959).
14. D. O. Edwards, J. L. Baum, D. F. Brewer, J. G. Daunt, and A. S. McWilliams, in *Proc. 7th Intern. Conf. on Low Temp. Phys. 1960*, North-Holland, Amsterdam, (1961), p. 610.