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POSITIVE-ION MOBILITIES IN LIQUID HELIUM I†

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Seven distinct positive-ion mobilities have been found in liquid helium I at 4.2°K. These mobilities correspond to the plateau regions of a mobility discontinuity curve similar to those discovered earlier in liquids helium II, argon, and nitrogen. A semiempirical equation is given which relates the mobilities corresponding to the plateau regions to a set of integers.

Discontinuities in the positive-ion mobility in liquids helium II,[1,2] argon, and nitrogen[3] have been previously observed. Discontinuities in the negative-ion mobility have also been observed[2] in liquid helium II at 2.00°K. It seemed likely that such mobility discontinuities should exist in classical liquid helium I since they are present in liquids argon and nitrogen.[4] This Letter reports the positive results of such a search in liquid helium I at 4.2°K. In addition a semiempirical relation is reported which relates the various constant-mobility regions to a hypothetical integer Δn.

The apparatus and technique used for this study are essentially the same as those described in Ref. 3, with the noted exceptions. The drift space in the time-of-flight spectrometer was decreased to 0.2 cm to allow higher electric fields to be reached at moderate voltages and a very accurate regulated power supply supplied the drift-space potential. A much faster dc electrometer-amplifier was used in the current-detection circuit. The output signal was averaged with a Princeton Applied Research waveform eductor (PARWE) to remove noise and then read out with an accurate oscilloscope. Occasionally as many as three different times of flight were detected on the same oscillogram after being averaged by the PARWE, but only one or two were observed on most oscillograms.

A considerable amount of data was taken in the 0–7000 V/cm range of electric field strengths so that a reasonable statistical analysis of data could be made. Seven distinct positive-ion mobilities were observed[5] at 4.2°K and 1 atm pressure. The results of the measurements are shown in Table I, tabulated versus the somewhat arbitrary integer Δn about which more will be said later in this Letter. The value of mobility $\mu = 5.37\times10^{-2}$ cm^2/V sec has been arbitrarily assigned the number $\Delta n = 0$ because of its excellent agreement with the very-low-field positive-ion mobility data reported by Meyer, Davis, Rice, and Donnelly[6] in liquid helium at 4.2°K and 1 atm pressure.

No attempt was made in this study to determine where the discontinuities occurred as a

Table I. Observed positive-ion mobility data for the mobility plateaus in liquid helium I at 4.2°K tabulated with respect to the integer Δn as determined from the experimental data. Errors represent the standard deviation from the mean mobility μ for each plateau.

Integer Δn	Mobility μ for the Δnth plateau (10^{-2} cm^2/V sec)
−3	6.47 ± 0.10
−2	6.08 ± 0.12
−1	5.70 ± 0.11
0	5.37 ± 0.07
+1	5.08 ± 0.09
+2	4.77 ± 0.08
+3	4.54 ± 0.17

function of electric field, but rather to obtain reliable values for the mobilities in the plateau regions.

Those experimental data for the average drift velocity of the ions varied linearly with the electric field in each of the various constant-mobility regions. The linear-correlation coefficient was greater than 0.999 as determined by linear regressions and all straight lines converged to the origin within experimental error.

It has been suggested that these mobility "steps" may be due to changes in the effective hard-sphere cross section of ions.[3] The clustering of atoms about an ion because of the inhomogeneous field of the charge has been studied extensively in gases.[7] It is well known that such clustering also occurs about positive ions in liquids. Atkins[8] was the first to study theoretically the electrostriction caused by an ion in liquid helium. Particularly noteworthy studies of a more experimental nature concerning such ion complexes in liquid helium are those of Dahm and Sanders[9] and Parks and Donnelly.[10] These studies were primarily aimed at liquid helium II but do contain considerable reference to helium I. Results of such studies indicate that the ion cluster may consist of from 40 to 100 helium atoms with a radius of 5.8 to 6.7 Å.

These data for ion size support an atomic close-packing hypothesis for an ion cluster. In ideal close packing one would expect the radius of the ion R_i to be proportional to the cube root of the number of particles N in the cluster. Thus $R_i \simeq R_a N^{1/3}$ where R_a is approximately the atomic radius of He$_4$. Since the kinetic-theory equation for the mobility is inversely proportional to the effective hard-sphere collision cross section $\pi(R_i + R_a)^2$, one would expect $\mu^{-1/2} \propto R_a(1+N^{1/3})$ where μ is the ion mobility. If we assume that at low fields the basic cluster size is N_0 and N changes by some integral amount ΔN then $\mu^{-1/2} = KR_a[1+(N_0+\Delta N)^{1/3}]$ where K is the proportionality constant. If $N_0 \gg \Delta N$ then $\mu^{-1/2} \simeq KR_a[1+N_0^{1/3}+\Delta N/3N_0^{2/3}]$. Figure 1 shows the data of Table I presented as $\mu^{-1/2}$ vs Δn where it is assumed that ΔN in the above equation is related to the integer Δn by $\Delta N = \Delta n N_1$. N_1 represents some hypothetical unit change in cluster size which is unknown. The straight line in Fig. 1 represents the least-squares linear fit to the data points. The linear fit to the data points is excellent and yields the empirical relation $\mu \simeq (4.31 + 0.13\Delta n)^{-2}$ where the units for μ are the same as those on the graph.

From the slope of the line it is possible to estimate values of N_1 by assuming values of N_0. If $N_0 = 40$ then $N_1 \sim 4$. On the other hand if N_0 is as much as 100 then $N_1 \sim 9$. By extending the kinetic-theory equation for mobility of ions in gases[11] to those atomic densities of liquid helium it is possible to calculate values for N_0 and N_1 directly from those mobility data of Table I. These calculations yield $N_0 \sim 99$ and an average $N_1 \sim 10$. The agreement between this value for N_1 and that value for N_1 computed from the experimental slope above with $N_0 = 100$ is encouraging.

It should be noted that Dahm and Sanders[9] have shown that the mobility for positive ions in liquid helium I at low electric fields can be approximated by a Stokes's law type equation for ion mobility. This type of mobility equation is known to be valid when the mean free path L of the ion is much less than the radius of the ion R_i. At the other extreme limit when $R_i \gg L$, the kinetic-theory mobility equation is known to apply. In liquid helium it would appear that the radius of the ion may be comparable with the mean free path. In this transition region it should be possible for both the kinetic-theory mobility equation and the Stokes's law mobility equation to approximately fit the experimental data for mobility. These two possible equations for mobility are neither contradictory nor inconsistent since

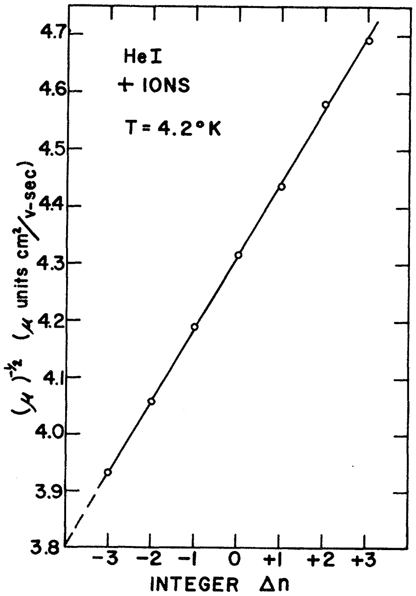

FIG. 1. Experimental data for the positive-ion mobility plateaus in helium I at 4.2°K presented as $\mu^{-1/2}$ versus the integer Δn. The ratio of the slope to the ordinate intercept yields 0.0297 and the slope is 0.128.

the conditions under which each applies merge continuously from one into the other.[12]

Regardless of the exact form for the mobility equation the foregoing discussion does not invalidate the close-packed cluster ion hypothesis as a possible explanation for these mobility "steps." Furthermore, Fig. 1 does tend to add credibility to this hypothesis even though there may be other equally credible explanations for the excellent data fits. However, it is clear that more experimental and theoretical work will be needed for a complete understanding of these phenomena.

Finally it should be remarked that those data for the positive-ion mobility "steps" in liquids argon and nitrogen[3] can also be represented by a graph similar to Fig. 1. It also appears, using data extrapolated from published graphs,[1,2] that those data for mobility "steps" in liquid helium II may be consistent with such a representation. Thus there may be more of a relationship between the mobility "steps" in the classical cryogenic liquids and the mobility "steps" in liquid helium II than has been previously suspected.

Note added in proof—Following submittal of this paper and long after completion of the research, there appeared a Letter by Schwarz[13] reporting an unsuccessful search for discontinuities in mobilities of ions in liquid helium II. The present work as well as earlier studies by this author[3] deal primarily with nonsuperfluid liquids so that field strengths and drift velocities are orders of magnitude different from Schwarz's. Schwarz in fact does not criticize the work in the nonsuperfluid domain. The discontinuities have been observed by the present author in Ar and N_2 as well as in He I with two completely different experimental facilities, spread over a time interval of nine years. The variations in experimental parameters made in an effort to expose the possibility of spuriousness are too great to recount in this Letter. The variations not only failed to falsify the discontinuities but established uniquely characteristic behavior patterns, different for each liquid. Finally, the present paper does not deal with variations correlated with the electric field strength.

In Schwarz's list of errors inherent in mobility measurements, some are confined to superfluids and hence are inapplicable here. Space-charge buildups would seem to round off discontinuities rather than accentuate them as Schwarz hypothesizes. Frequency dependences in the dc electrometry are difficult to envisage in the range from dc to 10 kHz with the electrometer-amplifiers used; the author has long since explored and eliminated possible irregularities of this type. Two separate amplifiers were used in this work—both with band passes from 50 to 300 kHz.

It must be concluded that the present paper is not directly comparable with Schwarz's.

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[1] G. Careri, S. Cunsolo, and P. Mazzoldi, Phys. Rev. Letters 7, 151 (1961), Phys. Rev. 136, A303 (1964).

[2] L. Bruschi, P. Mazzoldi, and M. Santini, Phys. Rev. Letters 17, 292 (1966), and Phys. Rev. 167, 203 (1968).

[3] B. L. Henson, Phys. Rev. 135, A1002 (1964).

[4] It should be noted that L. Bruschi and M. Santini, Rev. Sci. Instr. 41, 102 (1970), have recently confirmed the mobility "steps" reported by Henson (Ref. 3) in liquid nitrogen.

[5] Preliminary results were reported in B. L. Henson, Bull. Am. Phys. Soc. 14, 196 (1969).

[6] L. Meyer, H. T. Davis, S. A. Rice, and R. J. Donnelly, Phys. Rev. 126, 1927 (1962).

[7] Such ion clusters have been extensively studied in gases. For a comprehensive survey see L. B. Loeb, *Basic Processes of Gaseous Electronics* (Univ. of California, Berkeley, 1955), pp. 118-129.

[8] K. R. Atkins, Phys. Rev. 116, 1339 (1959).

[9] A. J. Dahm and T. M. Sanders, Jr., Phys. Rev. Letters 17, 126 (1966).

[10] P. E. Parks and R. J. Donnelly, Phys. Rev. Letters 16, 45 (1966).

[11] H. S. W. Massey and E. H. S. Burhop, *Electronic and Ionic Transport Phenomena* (Clarendon Press, Oxford, 1952), p. 367.

[12] For a more complete discussion of this transition in gases see L. B. Loeb, *Basic Processes of Gaseous Electronics* (Univ. of California, Berkeley, 1955), p. 172.

[13] K. W. Schwarz, Phys. Rev. Letters 24, 648 (1970).