P. de Magistris, I. Modena and F. Scaramuzzi: DEPENDENCE OF IONIC MOBILITIES IN LIQUID He^3 ON TEMPERATURE AND DENSITY.

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P. de Magistris, I. Modena and F. Scaramuzzi: DEPENDENCE OF IO-
NIC MOBILITIES IN LIQUID He^3 ON TEMPERATURE AND DENSITY(x).

We have measured the mobility of positive and negative ions in
liquid He^3 as a function of the temperature and of the density of the li-
quid. Previous measurements had been performed by Meyer, Davis, Ri
ce and Donnelly[1] and in this laboratory[2,3], but they were not so ex-
tensive as those presented here.

We used two different techniques in performing the mobility mea
surements. The first technique - the same used to obtain the measure-
ments reported in ref. (2) and (3) - is based on the method devised by
Cunsolo[4] for He^4 and consists of the measurement of the time-of-flight
of ions between two electrodes, whose distance is known. Recently one
of us (F. S.) developed a new method based on the measurement of the
voltage-current characteristics of a diode in condition of complete space
charge limitation. This condition is defined by having zero electric field
at the ion source electrode. The integration of the Poisson's equation gi
ves the following simple relation :

$$ i = \alpha \, \mu \, V^2 $$

where i is the measured current, V the applied potential, μ the ionic mo
bility and α a coefficient depending on the geometry of the cell. This
method has been successfully used in the measurement of ionic mobilities
in He^4[5]. The latest measurements have been performed with this me-
thod, which is faster and simpler to use and gives smaller errors. The
time-of-flight method requires that the distance between two electrodes
be known. The complete space charge method requires, besides this, the
knowledge of the absolute values of the current and of the voltage. We ha
ve preferred to calibrate the complete space charge cell with respect to
one experimental point taken with the time-of-flight method: the agree-
ment for the other data is satisfactory. The electric fields used with the

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complete space charge method are rather lower than those used for the other method : they were never above 30 v/cm.

The technique used to increase the pressure in the experimental cell has been described in ref. (3). Briefly, it consists of a stainless steel Toepler pump (a bellows filled with mercury), which enabled us to reach pressures up to 25 Atm using not more than 2 lt NTP of He^3. Before entering the cell, the He^3 was passed through a trap at liquid He^4 temperature. A standard type of continuous He^3 refrigerator enabled us to reach the temperature of 0.39°K.

In figs. 1 and 2 the values of the mobility for positive and nega tive ions respectively are reported as a function of the density of the liquid, for different temperatures. For negative ions the vapour pressu re values are indicated by a dotted line (fig. 2). For positive ions this has not been made, as the vapour pressure mobilities had already been presented in ref. (3). Both the straight lines in fig. 1 and the curves in fig. 2 must be considered as a rough attempt to find an empirical best fit to the data for a fixed temperature. The errors reported in both figu res are not to be intended as statistical errors, but as a measure of the uncertainty with which the straight lines can be thrown through the expe rimental points (in both methods it is necessary to throw a straight li ne : see ref. (4) and (5)).

It is not possible at present to fit our experimental data in any satisfactory theory. We will limit ourselves to point out a few conside rations about the limits within which a classical model can fit the data. The classical approach to this problem can be summarized in the two formulas describing the motion of an extraneous particle in a fluid, due to Einstein

$$(1) \qquad \mu = \frac{e\,D}{k\,T}$$

and to Stokes-Einstein

$$(2) \qquad D = \frac{k\,T}{6\,\pi\,\eta\,R}$$

Combining them, we get the well known Walden's rule

$$(3) \qquad \eta\mu = \text{constant} ,$$

valid in the hypothesis that the radius of the particle is constant. Table I shows the values of $\eta\mu$ in arbitrary units calculated for positive and negative mobilities at vapour pressure as a function of temperature. From table I it appears that equation (3) is satisfied only for positive ions for temperatures down to ~ 1.4 K. The lack of data for η values under pressure doesn't allow us to extend this kind of computation to larger liquid densities.

Another quantity which can be calculated in the approximation of this model is the diffusion coefficient of the ion, D_{i+}, from equation (1). As already noted in ref. (3), D_{i+} exhibits the same temperature de-

pendence of the self diffusion coefficient down to $\sim 1.4^\circ K$ (the ratio of the absolute values D/D_{i+} is about 7). But the values at lower temperatures show a complete disagreement between the two diffusion coefficients. In particular in the D_{i+} curve there is not any minimum corresponding to that in the D curve.

<div align="center">TABLE I</div>

$\eta\mu$ product in arbitrary units for positive and negative ions computed from smoothed values of μ at vapour pressure and from the viscosity measurements by Betts et al. [6].

T	$\eta\mu_+$	$\eta\mu_-$
3.0	17.7	6.3
2.5	18.5	7.0
2.8	18.5	7.5
1.5	18.6	7.9
1.3	19.9	8.5
1.0	22.2	9.4
0.8	23.9	10.0
0.5	30.0	12.2

Referring to the density dependence of the mobility, we want to point out that even the data for the self diffusion coefficient under pressure given by Garwin and Reich[7] show a behaviour almost linear in the same range of densities as ours, like the mobility of positive ions (see fig. 1).

A simple model for the positive ion was given by Atkins[8], who computed the effective mass of the ion due to polarization forces. Though the model is very rough, we may suppose the radius of the ion to be the distance at which the pressure equals the melting pressure. This implies the picture of the "snow ball", or solid cluster ion. The values of R so calculated and those from the values of μ using the Stokes formula

$$R = \frac{e}{6\pi\eta\mu}$$

are reported in table II. The two sets of values are in good agreement as far as the magnitude is concerned, but the dependence in temperature is instead opposite.

To conclude, we want to point out that recently theoretical works appeared in which the motion of the ions is considered in systems made up of bosons[9] or fermions[10], but it is impossible to test these theories making use of our data, because they are valid only at temperatures

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lower than those we had to deal with. Work is in progress to extend the temperature range of our measurements.

TABLE II

Comparison between the Atkins radius (snow ball model) and the Stokes radius (see equation (5)), both in Å, as a function of temperature.

T	R(Atkins)	R(Stokes)
3. 0	4. 55	4. 78
2. 2	4. 95	4. 55
1. 6	5. 40	4. 53
0. 8	6. 05	3. 54

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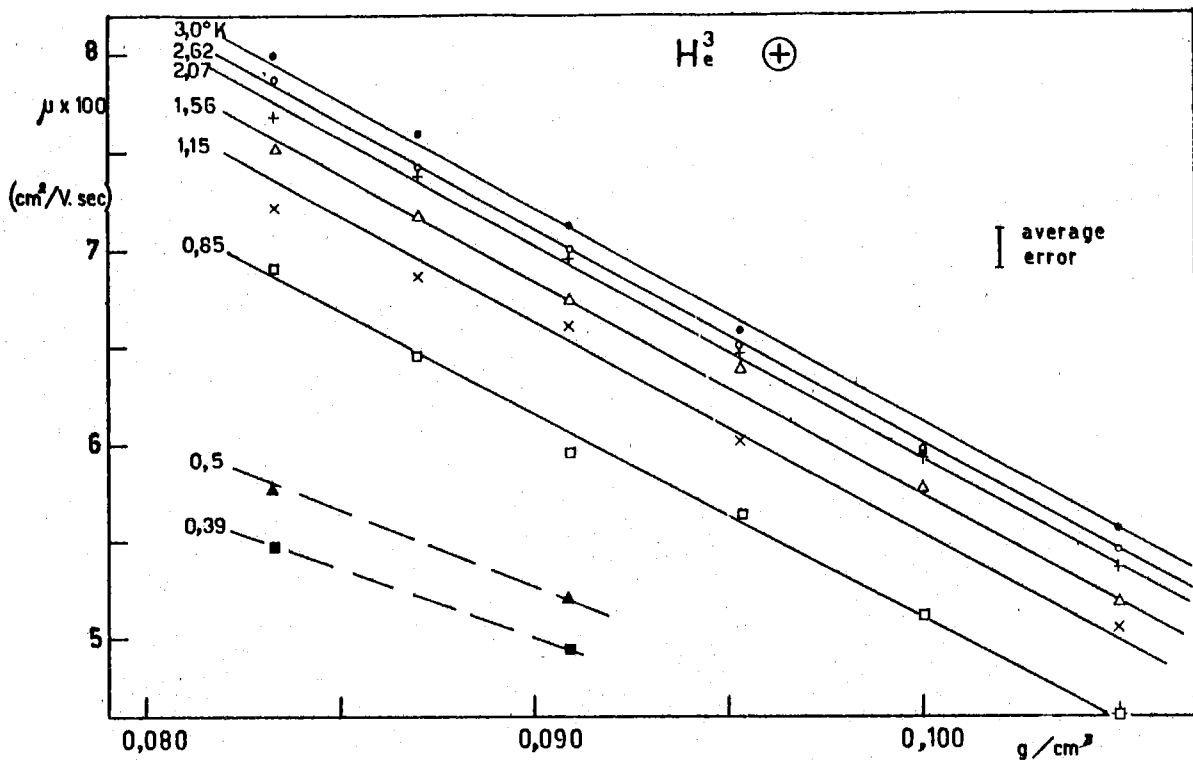(10) - R. C. Clark, Proc. Phys. Soc. 82, 785 (1963).

FIG. 1 - Positive ion mobility at different constant temperatures versus the density of the liquid.
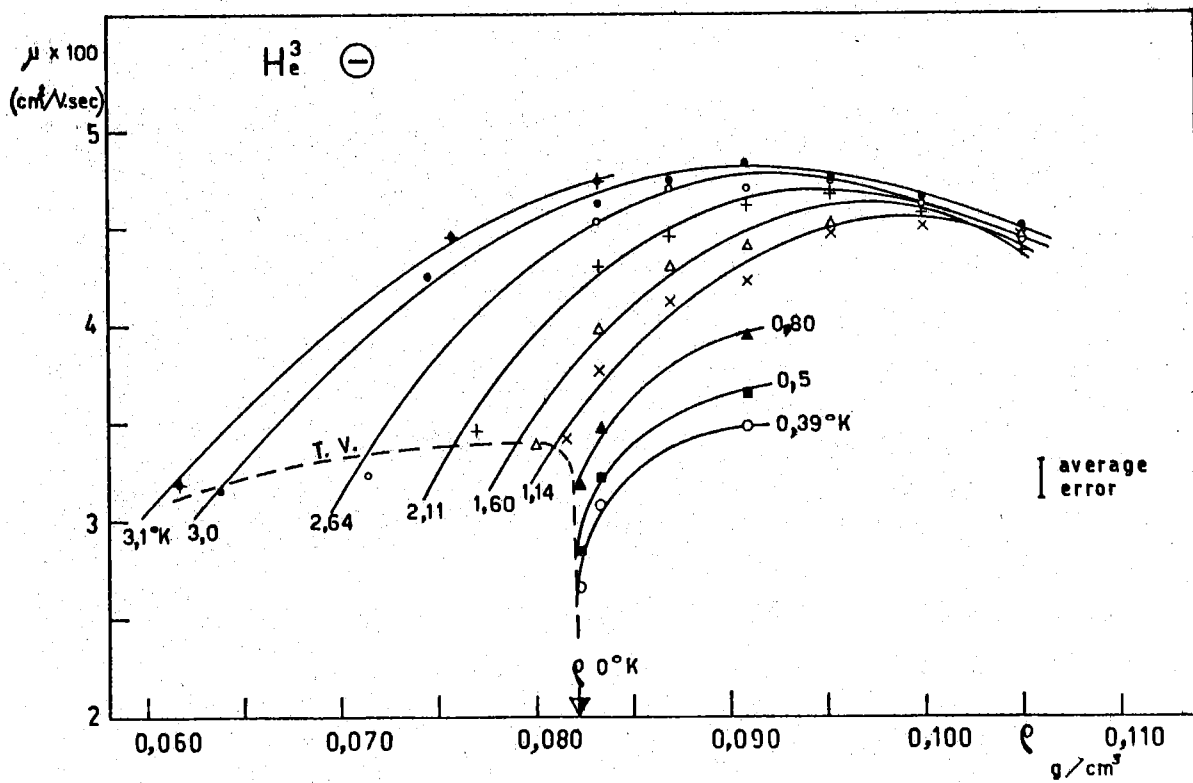

FIG. 2 - Negative ion mobility at different constant temperatures versus the density of the liquid. The dotted line shows the vapour pressure values.