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MOBILITY OF POSITIVE IONS IN He^3-He^4 *SOLUTIONS*

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A time of flight technique is used to measure the mobility of positive ions in He^3-He^4 solutions with He^3 content up to 29.0 mol.% in the temperature interval 1.40—2.15°K in fields up to 300 V/cm. It is shown that with increasing He^3 concentration, the mobility of the ions in superfluid helium decreases; this decrease is maximal in the region of low temperatures. The ion mobility connected with scattering by impurities is determined on the basis of the experimental data. The results are compared with the existing theoretical concepts.

THE mobility of positive and negative ions in liquid superfluid solutions is determined not only by the interaction with the rotons and phonons, as in pure He^4, but also by the interaction with the impurity excitations connected with the presence of He^3 in the solutions. The very first experiments performed with solutions of the helium isotope[1,2] have made it possible to observe this influence of He^3 and to obtain preliminary data on the magnitude of this effect. In this communication we present the results of experiments performed with solutions containing up to 29.0 mol.% He^3 in the temperature interval 1.40—2.15°K and in fields up to 200 V/cm.

The ion mobility was determined by a direct method[3,2] by measuring the time necessary for the ions to traverse the distance between two pairs of grids A_1A_2 and A_3A_4 (Fig. 1) which served as shutters. On each pair of grids there were applied simultaneously rectangular electric pulses, and the positive ions which were used in this investigation passed without obstacle through the entire apparatus only if the pulse polarity was suitable. The maximum current in this case is reached when the travel time of the ions between two shutters becomes equal to or a multiple of the period of the pulses. Naturally, when the frequency changes, maxima should be observed on the frequency dependence of the ion current, and from the positions of these maxima it is possible to determine the time necessary for the ions to traverse the distance between two shutters, and consequently to determine their mobility.

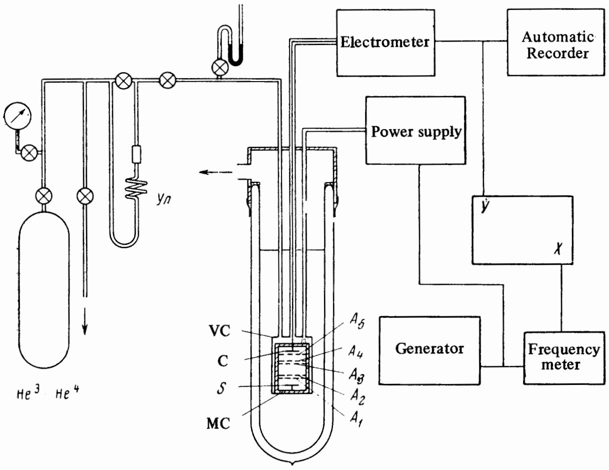

FIG. 1. Experimental setup: MC – measuring cell, VC – vacuum chamber, CT – carbon trap with filter, S – electron source, C – collector, A_1 – A_5 – grids.

The measuring cell MC (Fig. 1) located in a brass chamber VC, consisted of a source S and a collector C. The source was a molybdenum disc, coated with a layer of titanium tritide, emitting $\sim 10^7$ electrons per second with average energy 6 keV, and placed in the measuring cell.[1] The interaction of these electrons with the helium atoms led to the formation of ions in a layer $\sim 10\mu$ thick next to the source, the ions of appropriate polarity were directed by the homogeneous electric field to the collector. Five grids of copper-plated tungsten wire 15 μ thick, soldered with copper to molybdenum rings, were placed between the source and the collector. The rings had a diameter 15 mm and were also made of copper, the transparency of the grids being $\sim 90\%$. The two pairs of grids A_1A_2 and A_3A_4, as already noted, served as shutters, while the grid A_5 served as a screen. The distances between the grids were set by means of washers of organic glass and were as follows: $SA_1 = 2.0$ mm, $A_1A_2 = 0.50 \pm 0.02$ mm, $A_2A_3 = 2.50 \pm 0.02$ mm, $A_3A_4 = 0.50 \pm 0.02$ mm, $A_4A_5 = 2.0$ mm, and $A_5C = 2.5$ mm. The drift length, equal to the distance between the central points of the shutters, was thus 3.00 ± 0.05 mm. The volume of the measuring cell did not exceed 3 cm^3.

Teflon gaskets were used for the electrometric and the power leads; currents on the order of $10^{-15}-10^{-13}$ A were measured by an electrometer with a V2-5 dynamic capacitor and recorded with an automatic recorder. The frequency was measured by an electron-counting frequency-meter ChZ-3. The circuit made it possible both to record automatically the dependence of the ion current in the frequency, and to perform measurements point by point.

The procedure of working with charged particles in liquid helium consisted in the following. After the lowest temperature was reached in the cryostat (1.4—1.5°K), potentials of suitable polarity were applied to the grids of the measuring cell simultaneously with the condensation of the solution. The shutters remained open at the instant of condensation. Inasmuch as initially the liquid was lower than the source, the ions moved in the gas

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[1] The authors thank A. I. Shal'nikov for supplying the source.

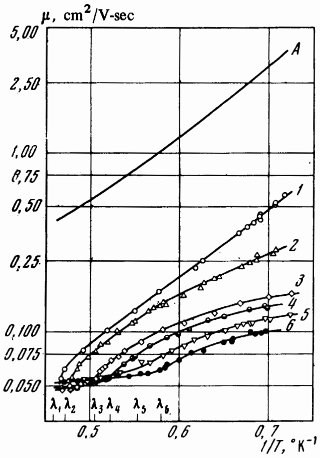

FIG. 2. Dependence of the mobility of positive ions in He^4 and in He^3 -He^4 solutions on the temperature. Curve 1 – He^4, solid line – data of [3,6], O – present work; curves 2 – 6 – solution He^3-He^4, with He^3 content 3.9, 13.0, 17.1, 23.7, and 29.0 mol.%, respectively. Curve A – calculated by formulas (2) and (3). λ_1 – λ_6 denote the temperatures of the λ transitions of He^4 in the corresponding solutions.

and the current was on the order of 10^{-10} A. At the instant when the liquid covered the source, the current dropped practically to zero, owing to the appearance of a liquid-vapor interface, and current appeared only when the liquid reached the collector. It was thus possible to trace the process of condensation and the instant of filling of the cell with the solution. The current in the liquid, at a field intensity 200 V/cm, is $\sim 10^{-12}$ A, i.e., smaller by two orders of magnitude than in a gas at the same field intensity.

The isotope mixture was condensed in the measuring cell through a carbon trap cooled with nitrogen, and a special filter with electrostatic cotton was used to prevent the entry of carbon dust particles. The temperature in the cryostat was maintained constant with an electronic stabilizer accurate to within 5×10^{-5} K.

The isotope mixtures were prepared by mixing pure He^3 and He^4. To exclude the influence of possible depletion of the light isotope, the concentration of the solutions was mounted to it during all the experiments by measuring the vapor tension[4].

After filling the cell with liquid, the constant voltage was disconnected from the shutters, rectangular pulses were applied, and the dependence of ion current on the pulse frequency was measured. These curves usually revealed three maxima, and it should be noted that the positions of the second and third maxima usually did not correspond to exact multiples of the frequency. This is possibly connected with the fact that the ratio of the widths of the shutters to the drift length A_1A_2/A_2A_3, which equaled 0.2 in our case, is not sufficiently small and part of the ion beam was cut off by the shutters.

Usually the time necessary for the ions to cover the distance between the shutters was determined from the position of the first maximum. The use of the second or third maximum for the same purpose has led to data that differ by not more than 5%. The scatter of the experimental data from experiment to experiment did not exceed 4%.

In the experiment aimed at studying the mobility, certain peculiarities were observed, to which attention was already called by Arkhipov and Shal'nikov[5]. The gist of these phenomena consist in the fact that if ions move in the liquid helium for a certain time under the influence of an electric field of suitable polarity, say positive ions, than when the direction of the electric field in the cell is reversed, the current of the negative charges reaches the maximum value not immediately but after an appreciable time. On the other hand, even when working during the course of the entire experiment with charges of the same polarity, the current does not remain constant at constant temperature and field intensity, but increases slowly, and this growth could reach 80—100% within the several hours required for the experiment.

It should be noted, however, that in spite of the dependence of the ion current on the prior history of the helium, the positions of the maxima on the plot of the current against the frequency, and thus also the determined value of the mobility, turned out to be insensitive to phenomena of this kind. This provides a sufficient guarantee that the indicated phenomena did not distort the results of the measurements of the ion mobility and the liquid helium, carried out with the aid of the customarily employed time of flight procedures with alternating current.

The results are shown in Fig. 2, which represents the mobility μ of the positive ions as a function of the reciprocal temperature in pure He^4 and in solutions He^3—He^4 containing 3.9 ± 0.2, 13.0 ± 0.2, 17.1 ± 0.2, 23.7 ± 0.2, and 29.0 ± 0.3 mol.% He^3 in fields up to 300 V/cm and in the temperature interval 1.40—2.15°K. The figure shows good agreement between the mobilities of the ions in pure He^4, measured in the present paper (circles), and the results of[3,6] (solid line). The presence of He^3 atoms greatly decreases the mobility of the ions, and this decrease is maximal in the region of low temperatures. Thus, for example, for a solution containing 29.0% He^3 at 1.4°K, the mobility is nearly one-sixth that of He^4. We call attention to the fact that for concentrated solutions, the mobility in the region of low temperatures is already weakly dependent on the temperature. The ion mobility in this region is determined apparently essentially by the scattering from the He^3 atoms.

Each μ(1/T) curve exhibits a kink connected with the transition through the λ point.

It should be noted that whereas in the He II region the presence of He^3 lowers the mobility of the ions, in the He I region the mobility of the ions in the solutions becomes somewhat larger than in pure He^4. This is connected with the fact that the mobility of the ions in liquid He^3 [7] is higher than in He I. Therefore the mobility of the ions in He I increases with increasing He^3 content.

To explain the results, we can use the model proposed by Arkhipov for the mobility of charged particles in liquid helium[8], based on the known concepts concerning the nature of positive and negative ions in liquid helium[9,10]. In this model, the ion is regarded as a sphere of radius R_i moving under the influence of the electric field $\mathscr{E}$ with velocity v in a sufficiently rarefied gas of phonons and rotons, so that the characteristic free paths are $l \gg R_i$.

For the decelerating force exerted on such a sphere by the excitation gas, Arkhipov obtained the following expression:

$$F = e\mathscr{E} = \frac{4\pi R_i^2}{3} v \int pn\, d^3p, \tag{1}$$

where p—momentum of the corresponding excitation, e—ion charge, n—distribution function normalized such that $\int nd^3p = N$, where N—number of excitations per unit volume. In the case of pure He^4, the integral should be taken respectively over the roton and phonon parts of the spectrum. At sufficiently high temperatures, when the influence of the phonon part of the spectrum can be neglected, the following expression is obtained for the mobility of the ions in superfluid helium:

$$\mu_r = \frac{3e}{4\pi R_i^2}\frac{1}{p_0 N_r}, \tag{2}$$

where $p_0 = 2.02 \times 10^{-19}$ is the zero-point momentum of the roton and N_r is the number of rotons per unit volume.

The radius R_i of the positive ion in the electrostatic model is approximately equal to[9]

$$R_i \approx \left[\frac{1}{2}\frac{N\alpha}{V_0}\frac{e^2}{p_m}\right]^{1/4}, \tag{3}$$

where $N\alpha = 0.124$—molar polarizability of the helium, V_0—molar volume, and p_m—solidification pressure of liquid helium.

Using (2) and (3) we can calculate the mobility of the ions in superfluid helium in the temperature region where the influence of the phonons can be neglected. The results of the calculations are shown in Fig. 2 (curve A). The calculated values of the mobilities are approximately six times larger than the experimental ones. Inasmuch as the electrostatic model leads to a discrepancy with experiment in the determination of the ion by only 20%[11], the main cause of the difference in the mobility between experiment and theory is apparently the fact that the model of elastic spheres used in this case is not suitable for a description of the interaction between an ion and a roton. In particular, it is probably necessary to consider not only the scattering of rotons by a sphere of radius R_i, but also by the layer of condensed liquid formed around the charged particles under the influence of the electrostatic attraction.

In He^3—He^4 solutions there is added to the roton and photon parts of the spectrum another part connected with the presence of impurity excitations:

$$E = p^2 / 2m_{\text{eff}},$$

where E—energy, p—momentum, and m_{eff}—effective mass of excitation. Integrating relation (1) over the impurity part of the spectrum, we easily obtain an expression for the mobility of the ions in the He^3—He^4 solutions, due to the scattering by the impurities:

$$\mu_3 = \frac{3e}{4\pi R_i^2}\frac{\pi^{1/2}}{2N_3(2m_{\text{eff}}kT)^{1/2}}. \tag{4}$$

Here N_3—number of He^3 atoms per unit volume, $m_{\text{eff}} = 2.35\, m_3$[12], where m_3—mass of the He^3 atom. Expression (4), obtained with the aid of (1), coincides with the formula for the mobility of the ions diffusing in the gas under the condition that the mass of the ion be much larger than the mass of the gas atoms, $M_i \gg m$[13].

The temperature dependences of μ_3 for He^3—He^4 solutions containing 3.9, 13.0, 17.1, 23.7, and 29.0 mol.% He^3, calculated using (4), are shown in Fig. 3. In the calculation, as in the case of pure He^4, the radius of the positive ion was calculated in accordance with (3), in which the experimental values of the molar volumes and solidification pressures of the He^3—He^4 solutions were substituted[14,15]; in addition, a correction for the finite dimension of the He^3 atoms was introduced in the cross section for the scattering of the ion by the He^3 atom.

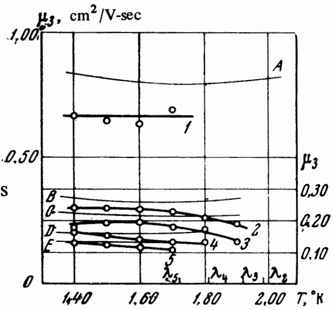

FIG. 3. Positive-ion mobility connected with scattering by He^3 atoms vs. temperature. Curves 1 – 5 – He^3-He^4 solutions with He^3 content 3.9, 13.0, 17.1, 23.7, and 29.0 mol.% respectively; curves A – E – calculation by means of formulas (3) and (4). Curves 1, A – scale on the left, curves 2 – 5, B – E – scale on the right.

Inasmuch as the influence of phonons on the mobility of the ions can be neglected in the investigated temperature region, the mobility of the ions in the solution can be represented in the form

$$\mu^{-1} = \mu_r^{-1} + \mu_3^{-1}, \tag{5}$$

where μ_r—mobility of the ions connected with scattering by rotons, and μ_3—ion mobility connected with scattering by impurities. Relation (5) makes it possible to obtain the value of μ_3 from the experimental data. Figure 3 shows the dependence of the ion mobility due to scattering by impurity He^3 atoms on the temperature, calculated in the same manner, for the same solutions (curves 1—5). As seen from the figures, the calculated and the experimental values of μ_3 approximately coincide. The value of μ_3 depends little on the temperature and, as expected, decreases with increasing He^3 content. In the calculation of μ_3 it was necessary to assume that the mobility of the ions μ_r, due to scattering by rotons in the He^3—He^4 solutions, coincides with the mobility of the ions, due to scattering by the rotons in the superfluid He^4. For weak solutions this assumption apparently is not subject to any doubt. The basis for the justification of such an assumption in more concentrated solutions may be the experimental data by measuring the density of the normal component and the velocity of fourth sound in He^3—He^4 solutions[12,16], which evidence of the applicability of the Pomeranchuk theory[17] to solutions with He^3 content at least up to 20%.

It is appropriate to note that on the basis of the foregoing the quantity $1/\mu_3 c$ in solutions should depend on the concentration c of the He^3 only to the extent that R_i changes as a result of the increase of the solidification pressure with increasing c, which in general agrees with the experimental data (Fig. 4).

The experiments will be continued in the region of lower temperatures.

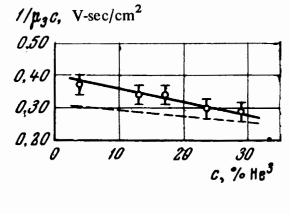

FIG. 4. Reciprocal mobility referred to a 1% concentration vs. He^3 concentration. Points – experimental, dashed curve – calculation by means of (3) and (4).

[1] F. Reif and L. Meyer, Phys. Rev. Lett. **5**, 1 (1960).

[2] Yu. Z. Kovdrya and B. N. Esel'son, ZhETF Pis. Red. **6**, 581 (1967) [JETP Lett. **6**, 90 (1967)].

[3] F. Reif and L. Meyer, Phys. Rev. **119**, 1164 (1960).

[4] B. N. Esel'son, Doctoral Dissertation, Khar'kov, 1957.

[5] R. G. Arkhipov and A. I. Shal'nikov, Zh. Eksp. Teor. Fiz. **37**, 1247 (1959) [Sov. Phys.-JETP **10**, 888 (1960)].

[6] S. Cunsolo, Nuovo Cimento **21**, 76 (1961).

[7] J. Modena, Proc. of the 8th Int. Conf. on Low Temp. Phys., London, 1963, p. 51.

[8] R. G. Arkhipov, Usp. Fiz. Nauk **88**, 185 (1966) [Sov. Phys.-Usp. **9**, 174 (1966)].

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

[10] R. A. Ferrel, Phys. Rev. **108**, 167 (1957).

[11] A. Dahm and T. M. Sanders, Phys. Rev. Lett. **17**, 126 (1966).

[12] V. N. Grigor'ev, B. N. Esel'son, V. P. Mal'khanov, and V. I. Sobolev, Zh. Eksp. Teor. Fiz. **51**, 1059 (1966) [Sov. Phys.-JETP **24**, 707 (1967)].

[13] B. M. Smirnov, Usp. Fiz. Nauk **92**, 75 (1967) [Sov. Phys.-Usp. **10**, 313 (1967)].

[14] E. C. Kerr, Low Temp. Phys. and Chem., Madison, 1957, p. 158.

[15] B. N. Esel'son and B. G. Lazarev, Dokl. Akad. Nauk SSSR **97**, 61 (1954).

[16] B. N. Esel'son, N. E. Dyumin, E. Ya. Rudavskii, and I. A. Serbin, Zh. Eksp. Teor. Fiz. **51**, 1064 (1966) [Sov. Phys.-JETP **24**, 711 (1967)].

[17] I. Ya. Pomeranchuk, ibid. **19**, 42 (1949).

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