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Charge movement in crystalline helium

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The mobilities of the carriers in crystalline helium are determined by investigating the diode volt-ampere characteristics and the temperature dependence of the currents induced by a tritium source. The diode current is found to be proportional to the square of the voltage in a comparatively narrow range of voltages and temperatures. In the range of high voltages, the form of the volt-ampere characteristics is determined by the dependence of the drift velocity of the charges on the electric field strength. For direct measurement of the carrier drift velocity, a three-electrode time-of flight method is employed. The temperature dependences of the positive and negative carrier mobilities are measured at different molar volumes. The dependence of the carrier drift velocity on the electric field strength is also measured. The data are compared with the results of existing theoretical studies.

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The first experiments on the movement of charges in solid helium were made by Shal'nikov[1] and were continued in Refs. 2 and 3. In these studies specially grown helium crystals served as the first object of the investigation. (An attempt at current measurement in polycrystalline samples obtained by the blocked capillary method showed that the currents in this case were not amenable to measurement.[4]) In Refs. 1–3, the helium crystals were grown from the liquid phase at constant pressure in a cylindrical glass container. The growth rate was determined by the temperature gradient along the container axis. The growth process could be monitored visually. The decrease in the induced diode current during the solidification process was the criterion of the quality of the crystal sample. Thus, in the solid phase, the current amounted at most to 2–3% of the current in liquid helium for the worst samples and reached 60% for the better samples. Usually, the better samples were obtained at a growth rate of ~ 5 μm/sec.

The described method of obtaining the crystals was used by Mezhov-Deglin in the study of the thermal conductivity of solid He^4.[5] The record values of thermal conductivity measured by him, up to 50 W/cm-deg, and the absence of an "annealing" effect, indicate that the samples obtained under these conditions were very nearly perfect single crystals. A direct verification of this fact was obtained in Refs. 6 and 7 by x-ray analysis.

The purpose of the present work was the study of the process of charge transfer in the He^4 crystals obtained by the described method. The research followed two basic directions:

1) measurement of the currents induced in the diode;

2) measurement of the charge velocity by a three-electrode time-of-flight method.

CONSTRUCTION OF THE APPARATUS

The construction of the apparatus used in the present research and the method of growing the crystals were described in detail in Refs. 2 and 5. The crystals were grown in containers of two types: glass (diameter 10 mm) and metal (diameter 40 mm). Inside each of them, depending on the goal of the experiment, was placed a diode or triode electrode system. The construction of the glass container with a small triode inside is shown in Fig. 1a. Rings of ferrochrome (alloy N47KhB) were

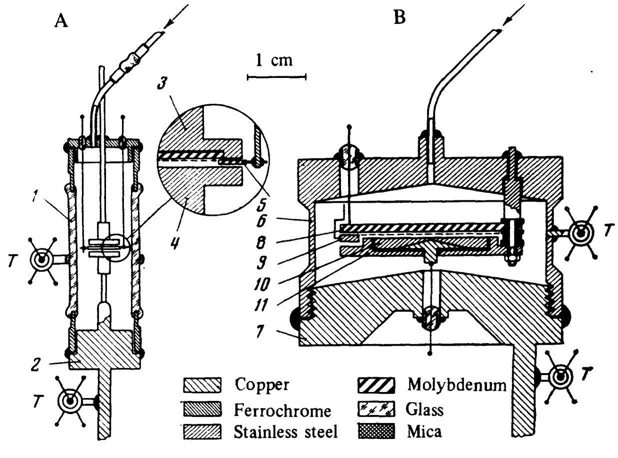

FIG. 1. Construction of containers and triodes. 1, 6—housing; 2, 7—bottom with cold conductor; 3, 8—β-active electrode; 4, 10—collector; 5, 9—grid; 11—guard ring; T—thermometers.

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soldered to the ends of a container of chemical glass (No. 23 or No. 29). A copper cold finger was soldered to the lower part of the container. The conducting ends and a filling capillary passed from above through glass insulators through the ferrochrome cap. The middle section of the container was tightly wrapped by a copper ring to which one of the thermometers was soldered. A second thermometer was soldered to the copper cold finger. The carbon thermometers were prepared in the form of thin disks cut from Allen-Bradley radio resistors. Three plane-parallel electrodes were placed in the center of the container. A molybdenum disk of 5 mm diameter was pressed into the upper electrode, which was of stainless steel of 8 mm diameter. The outer surface of the molybdenum was covered by a layer of titanium tritide, which served as a source of ionizing radiation. The electrons emitted by the source, having a maximum energy of ~ 18 keV, ionized an adjacent layer of helium ~ 10 μm thick. The charges throughout the rest of the interelectrode space could be of only one sign, corresponding to the sign of the potential of the upper electrode. The lower electrode (collector), with a diameter of 8 mm, was also prepared of stainless steel. A grid was placed between the source and the collector. A fabricated klystron mesh-washer of molybdenum of thickness 0.1 mm and diameter 6.5/3.5 mm was used in the small triode. On this washer was stretched a tungsten wire of diameter 8 μm, forming a one-dimensional lattice with a period of 30 μm. The distance between the grid and the collecter amounted to 280 μm and between the source and the grid—50 μm.

The construction of the metallic container and of the large triode is shown in Fig. 1b. The container housing, with inside diameter 40 mm, was made of stainless steel. A copper bottom with a cold finger was screwed into the lower part of the housing. The joint was soldered with soft solder. A copper wire passed through a seal in the wall of the container, establishing thermal contact between the inner volume of the container and one of the thermometers. A second thermometer was soldered to the cold finger. The upper β-active electrode was made of molybdenum in the form of a disk with diameter 31 mm. A large part (diameter 25 mm) of the lower surface of the disk was covered with a layer of TiT_2. A nickel grid 2 μm thick and with 30×30 μm mesh served as the second electrode. The grid was stretched on a steel ring with inside diameter 25 mm. The transparency of the grid was ~ 70%. A stainless steel collector was cemented in a guard ring with the help of polymerized resin (Stycast 126). The gap between the collector and the ring amounted to 0.2 mm. The system of electrodes was assembled on three pins fixed to the housing of the container. The electrodes were isolated by mica gaskets, which simultaneously fixed the spacing between them. The gap between the grid and the collector amounted to 300 μm and between the source and the grid—100 μm.

The small and large diodes differed from the triodes described above only in the absence of the grid.

DIODE CURRENTS IN SOLID He

In the study of the diode volt-ampere characteristics of the diodes, we were primarily interested in the possibility of using them for quantitative measurement of the mobility of the carriers (or, simply, ions)1) in solid helium. The corresponding electrodynamic problem was studied by J. J. Thomson,[8] who showed that, in the case of a plane-parallel diode, filled with a material, a thin layer of which is ionized near one of the diode electrodes by the radiation source, the initial segment of the volt-ampere characteristic is described by the formula

$$i=9\mu U^2 S/32\pi L^3, \tag{1}$$

where i is the current of the diode, U is the difference of potentials between the electrodes, S is the area of one of the electrodes, L is the distance between the electrons, and μ is the carrier mobility.

The distribution of the electric field along the x axis, perpendicular to the plane of the electrons, has the form

$$E(x)=\left(\frac{8\pi i x}{\mu S}\right)^{1/2}=\frac{3}{2}\frac{U}{L}\left(\frac{x}{L}\right)^{1/2}. \tag{2}$$

The relations (1) and (2) are valid upon satisfaction of the following conditions: a) $\delta/L \ll 1$, δ is the thickness of the ionized layer; b) charges of only one sign are present in the entire diode space (with the exception of the ionized layer); c) $i/I \ll 1$, where I is the saturation current of the diode; d) $v/E=\mu=\text{const}$, where v is the drift velocity of the charges. As is seen from formula (1), the current in the case considered does not depend on the intensity of the ionization source.

Thus, by measuring the volt-ampere characteristic in the region where $i\propto U^2$, and knowing the parameters of the diode, we can calculate the mobility μ. The described method has been used by Sai-Halasz and Dahm[9,12,13] for the determination of the mobility of ions in solid helium.

The results of our measurements of negative currents in solid helium in the small diode ($L=0.1$ cm, $S=0.2$ cm^2, $I=3.6\times 10^{-9}$ A) are given in Fig. 2. The He^4 crystal was grown under a pressure of 38 atm. For control of the quality of the crystal during its growth, the current was recorded at a constant voltage of 2 kV. At temperatures below 1.72 K, the current was measured at several values of the voltage in the range 0.6–4.0 kV, while the crystal was slowly cooled. No measurements were made at smaller voltages in these experiments for the following reasons. First, as seen from Fig. 2, the current in the solid phase falls off exponentially with decrease in the temperature, so that at small voltages, the temperature interval in which the values of the current lie in the measurable range $>10^{-14}$ A turns out to be too small. Furthermore, in repeated measurement of the volt-ampere characteristics at constant temperature, we encountered the phenomenon of hysteresis, which became more significant the smaller the measured currents. The measurements were already ambiguous at currents $<10^{-13}$ A. Similar phenom-

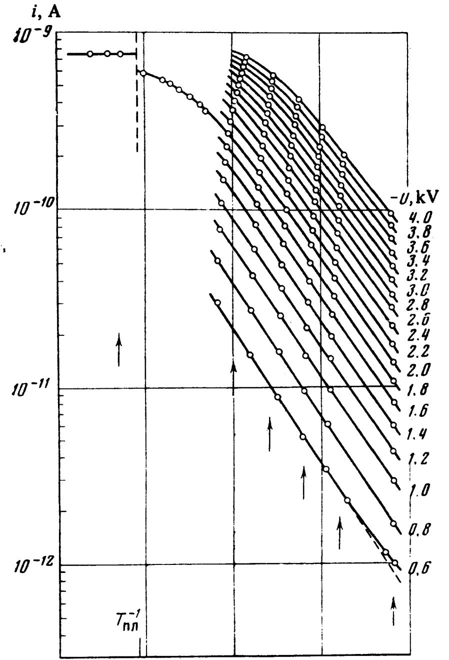

FIG. 2. Temperature dependence of the negative current in a diode at different voltages on the crystal at a pressure of 38 atm. The arrows indicate the temperatures at which the volt-ampere characteristics of Fig. 3 are constructed.

ena were also observed by the authors of Refs. 1 and 10. The reason for the hysteresis could be, for example, the capture of ions by different defects of the crystal and their accumulation in the volume of the sample.[10] However, we have also observed hysteresis in the volt-ampere characteristics in measurement of the weak currents even in liquid helium, where a similar explanation is obviously inapplicable. Since this question has not been considered specially in our work, the nature of the hysteresis remains unclear to us.

The temperature dependences of the current at different voltages were measured by us in several crystals, grown at a pressure of 38 atm, for charges of both signs. The results of the measurements of the positive currents are similar to the results shown in Fig. 2. The scatter of the data for the currents of one polarity from crystal to crystal does not exceed 10%.

A characteristic family of curves of the dependence of the square root of the diode current on the voltage at different temperatures, obtained from the experimental results given in Fig. 2, is shown in Fig. 3. In contrast with the results of Refs. 9, 12, 13, not one of the curves is described by Eq. (1) over the entire voltage range of 0.6–4 kV. At the beginning of each volt-ampere characteristic (except the one at $T = 2.14$ K, which refers to liquid helium near solidification), a region of quadratic variation is observed, and the maximum value of the voltage at which $i^{3/2} \propto U$ still holds decreases with decrease in the temperature. At currents that are larger than 4×10^{-10} A, the current growth continues on the curves corresponding to the temperatures 1.67 and 1.56 K, but more slowly than follows from Eq. (1). In the same way, Eq. (1) is not satisfied in the case of the liquid phase ($T = 2.14$ K) over the entire range of voltages studied.

On the curves corresponding to the temperatures 1.56–1.28 K, the initial segment $i^{1/2} \propto U$ at voltages of 1.5–2 kV transforms into a region characterized by a derivative that increases like $\partial i^{1/2}/\partial U$.

To explain the reasons for such a significant difference of the volt-ampere characteristics from the theoretical expressions, let us consider to what degree the conditions at which formula (1) is valid are satisfied in our experiments.

As was noted previously, the ionizing β radiation penetrates not more than 10^{-3} cm into the bulk of the helium if the distance between electrons is $L = 10^{-1}$ cm, i.e., in our case, $\delta/L \ll 1$. It is necessary to note here that the β radiation is not the only source of interelectrode ionization. The interaction of the tritium-decay β electrons with the material of the active layer and the substrate leads to the appearance of x radiation, the spectrum of which consists of the K line of titanium (4.5 keV), the L line of molybdenum (2.3 keV) and a bremsstrahlung background (2–12 keV). The x rays interact very weakly with the helium; however, they can generate the emission of photoelectrons from the surface of the collector with energies that are sufficient for ionization of the helium in a thin layer near the surface of the metal. The current passing through the diode will in this case be due to the motion of both the charges of the chosen sign to the collector as well as the charges of the opposite sign in the opposite direction. In special control experiments it was established that in the diodes studied by us the intensity of the ionization process near the collector amounted to no more than 0.1% of the intensity of ionization near the active electrode.

Thus, both the first necessary condition and the second, which requires the presence of charges of only one sign in the volume of the diode, are satisfied in our case with sufficient accuracy.

At the diode saturation current $I = 3.6\times10^{-9}$ A (measured in an ionization chamber), the ratio i/I for the volt-ampere characteristics shown in Fig. 3 obviously ceases to be sufficiently small at currents greater than

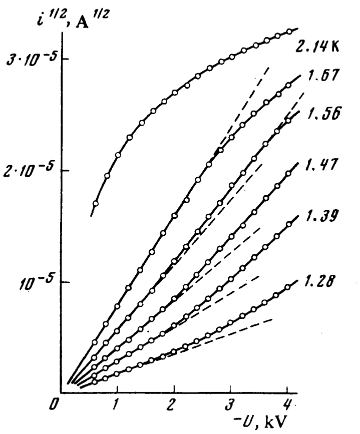

FIG. 3. Dependence of square root of the diode current on the voltage for negative ions at different temperatures in liquid and solid He^4 at a pressure of 38 atm. The points were obtained from the results of the experiment shown in Fig. 2.

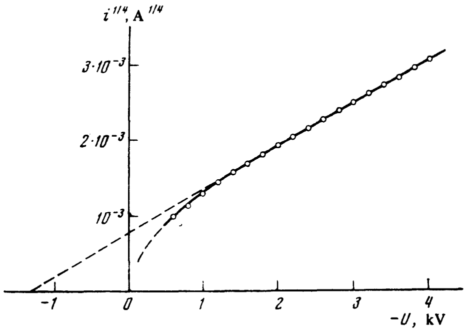

FIG. 4. Dependence of $i^{1/4}(U)$ for negative charges in a crystal at $T = 1.28$ K. $p = 38$ atm.

4×10^{-10} A. Recombination of charges into an ionized layer now becomes important here, and leads to a slower than quadratic dependence of the current on the voltage in this region.

It is difficult to explain the increase in the curvature of the characteristics $i^{1/2}(U)$, observed at voltages exceeding 1.5–2 kV, without invoking additional considerations. The assumption that in this region of voltages the drift velocity of the ions is not proportional to the electric field intensity is apparently more natural. However, it is not possible to carry out a quantitative analysis of the volt-ampere characteristics without actual knowledge of the dependence $v(E)$. We have succeeded in doing this only on the basis of the results of measurements of the dependence of the drift velocity of ions on the field strength, obtained by the time-of-flight method. As it turned out, in weak fields ($E < 10^4$ V/cm), the drift velocity is proportional to the field strength. In strong fields, the dependence of the drift velocity on the field strength has the form

$$v=\alpha(E+E_0)^3, \tag{3}$$

where α and E_0 are parameters that are independent of E.

Calculations similar to those of Ref. 8, with account of (3), lead to the result that at high voltages the volt-ampere characteristic of the diode has the asymptotic form

$$i=625\alpha(U+E_0L)^4S/4096\pi L^5. \tag{4}$$

Figure 4 shows the already considered volt-ampere characteristic at 1.28 K, plotted in the coordinates $i^{1/4}(U)$. At voltages above 1.2 kV, the dependence $i\propto(U+\text{const})^4$ is observed, in complete correspondence with (4). The value $E_0 = 1.3\times10^4$ V/cm, calculated by means of (4), agrees in order of magnitude with the results of the time-of-flight measurements at a pressure of 36.6 atm and temperature 1.43 K.

It should be noted that an increase in the slope of the characteristics $i^{1/2}(U)$ was observed by the authors of Refs. 12 and 13, only in certain crystals (which, in their opinion, were distinguished by a large dislocation density) as was attributed to the effect of the space charge, which arises as a result of the capture of the ions by the dislocations, whose charge density depends on the external field strength. However, the agreement of the experimental dependence $i(U)$ with Eq. (4) in strong fields indicates the absence of charge accumulation in the volume of the sample in our experiments.

The field strength at which the transition of the volt-ampere characteristics from region (1) to region (4) takes place decreases with decrease in the temperature. At the same time, because of the significant decrease in the current upon decrease in the temperature, the field strength at which the phenomenon of hysteresis does not affect the measurements, increases. As a result, the range of field strengths in which it is really possible to observe the dependence $i\propto U^2$ becomes narrower with decreasing temperature. This circumstance also determines the natural limit of possibility of calculating the mobility from the diode volt-ampere characteristics.

In order to eliminate the difficulties associated with the measurement of small currents, we used a large diode with the following parameters: $L = 4.1\times10^{-2}$ cm, $S = 3.3$ cm^2, $I = 9.2\times10^{-8}$ A. Figure 5 shows the temperature dependences of the positive and negative diode currents at a constant voltage of 300 V for two He^4 crystals. The first crystal, grown at a pressure of 32.4 atm, was gradually cooled to a temperature of 1.36 K. During the time of measurement of the currents, the temperature of the sample was specially stabilized to within 5 mK. The crystal was then annealed for 10 h at a temperature of 1.60 K (the melting temperature was 1.85 K), after which a second series of measurements was carried out in the temperature range 1.50–0.64 K. A third series of results in Fig. 5 refers to another crystal, grown at the same pressure and not subjected to annealing. The results of all the measurements differed at the lowest temperatures by not more than 20%.

In the high-temperature range, the dependence of the currents of both signs on the temperature has the form

$$i\propto\exp(-\varepsilon/T).$$

The values of the coefficient in the exponential for the positive and negative currents are respectively

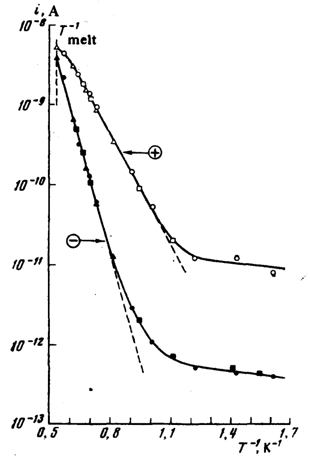

FIG. 5. Temperature dependence of positive and negative diode currents at a voltage of 300 V for two crystals of He^4 at a pressure of 32.4 atm; △, ▲—crystal No. 1 before annealing; □, ■—crystal No. 1 after annealing; ○, ●—crystal No. 2 without annealing.

$$\varepsilon_+=11.6\text{ K},\quad \varepsilon_-=21.1\text{ K}.$$

At temperatures $\sim 0.5\, T_{\text{melt}}$ the character of the temperature dependence of the current for ions of both signs changes qualitatively. Similar changes were observed previously[2,9,12,13] for crystals of different density. A conclusion was drawn on this basis as to the difference in the mechanism of motion of charges of both signs at "high" and "low" temperatures.[9,12,13]

However, it should be noted that the volt-ampere characteristics measured by us in the given crystals in the range 100–800 V were similar in form to that shown in Fig. 3, and in the case of negative changes they already differed significantly from quadratic at temperatures below 1.2 K. For this reason, we cannot deduce that any qualitative change occurs in the character of the ion mobility in the range of low temperatures on the basis of the observed temperature dependences of the currents. According to the results of the time-of-flight measurements (see Fig. 11 below) in fields $\sim 10^4$ V/cm, which correspond to a diode voltage of 300 V, at temperatures ~ 1 K and below, the drift velocity is not proportional to the field strength, as is undoubtedly reflected in the temperature dependence of the current.

Moreover, the change in the character of the dependence of the current at low temperatures can be due to an entirely different reason, namely, the ionization of the helium near the collector, which, as we have already mentioned, can take place as a consequence of the x-radiation of the source. Several results obtained by us in the measurement of the triode currents in solid helium indicate the important role of the process mentioned in the low-temperature region. The collector current i of the triode was measured as a function of the voltage U_1 between the source and the grid at a constant voltage U_2 between the grid and the collector. We consider the case $U_2<0$ for definiteness. The direction of the field E_1 in the space between the source and the grid, which acts as an electric gate, determines the sign of the charges advancing from the source to the grid. It is obvious that, in the absence of ionization between the collector and the grid, only negative charges can proceed to the collector and only if the triode is open ($U_1<0$). The penetration of the field E_2 into the gate can be the reason for a certain small collector current at $U_1=0$, whose value characterizes the effectiveness of the action of the gate. However, in the case of a closed triode ($U_1>0$), this current should be zero.

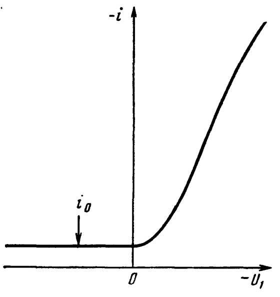

FIG. 6. Schematic dependence of the triode current on the voltage between the source and the grid at constant voltage between the grid and collector in He^4 crystals. The case of negative currents is considered ($U_2<0$); $T=$const.

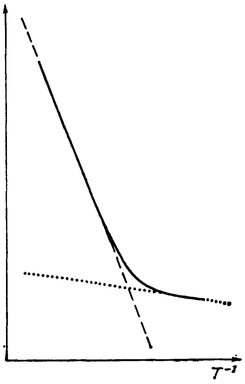

FIG. 7. Schematic temperature dependence of currents in open and closed triodes in He^4 crystals. $U_2=\text{const}<0$; solid line—i ($U_1=\text{const}<0$), dotted line— $-i_0$ ($U_1\geqslant 0$), dashed line—$i-i_0$.

In actuality, the dependence of the collector current of the triode on the voltage U_1 appears qualitatively in the following fashion (Figs. 6 and 7). The current i_0 in the closed triode differs from zero, does not depend on the value of the blocking voltage U_1 over a wide range of voltages, and decreases very slowly upon a decrease in the temperature. In the open triode, the current i is a monotonically increasing function of the voltage U_1 and at constant U_1 and U_2 it changes with decrease in temperature in a fashion similar to the diode current.

The fact that the collector current in the closed triode differs from zero and does not depend on the value of the blocking voltage can be explained only by the presence of ionization in the space between the grid and the collector and by the flow of charges in the collector-grid circuit (Fig. 8, I). The character of the temperature dependence $i_0(T)$ is evidently a consequence of the closeness of the current i_0 to saturation which, in view of the weakness of the ionization near the collector, sets in at comparatively low voltages U_2. In such a case, the collector current in the open triode is a superposition of two currents, due to the opposing motions of the positive and negative charges (Fig. 8, III).

Thus, in our case ($U_2<0$), the temperature dependence $i(T)|_{U_1=\text{const}<0}$ at high temperatures, where $i\gg i_0$, is determined essentially by the mobility of the negative charges; in the low temperature region ($i\approx i_0$) the current is practically entirely due to the passage through the crystal of positive charges only, and its temperature dependence is determined by the factors that prevail in the region near saturation.

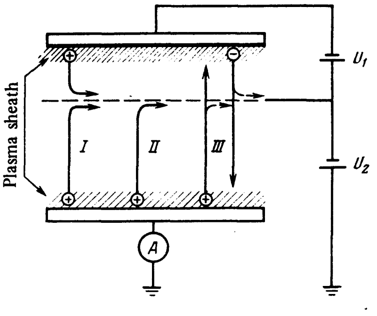

FIG. 8. Passage of charges through closed (I, II) and open (III) tridoes in the presence of additional ionization of helium near the collector. $U_2<0$; I—$U_1>0$, II—$U_1=0$, III—$U_1<0$.

The discussions given above are valid also in the case of the diode; however, it is not possible in principle to carry out the corresponding analysis on the basis of the results of measurements of the diode currents.

TIME-OF-FLIGHT MEASUREMENT OF THE ION DRIFT VELOCITY

A three-electrode time-of-flight method of measurement of the mobility of the carriers in solid helium has been used previously.[14] As it turned out, the mobility for charges of both signs near the melting curve amounted to $\sim 10^{-5}$ cm^2/sec-V. However, it has not been possible to measure the temperature dependence of the mobility, since the indicated values turned out to be extremely small in this procedure.

For the determination of the very small drift velocities of the charges in solid helium, it proved to be convenient to use the time-of-flight method,[15] based on measurement of the time of growth of the collector current in the triode.

The triode used in the present research was the same as in previous experiments[14,15] (Fig. 1a). In addition, to increase the collector current, the measurements were carried out in a triode of large diameter, located in a metallic container (Fig. 1b). The construction of the triode allowed us to measure directly the time of passage of charges of given sign through the drift space between the grid and the collector, and thus determine their drift velocity.

The procedure of measurement of the drift time is illustrated in Figs. 9a and b. The electric circuit is shown in Fig. 8. Before beginning the measurement, a constant voltage U_2 was applied between the grid and the collector. At the instant of time $t=0$, a voltage U_1 was applied between the source and the grid, of the same polarity as U_2. During a certain time corresponding to the passage of the front of charges from the ionized layer to the grid, current is absent. The penetration of charges through the grid into the drift space is accompanied by a collector-current growth that is linear with the time. This growth ends at the instant of arrival of the front at the collector. The time of buildup of the current from zero[2] to a constant value was taken as the time of flight τ.

The considered process was recorded as a function of the quantity and time of growth of the current by an electrometric amplifier assembled around the operational amplifier "Analog Devices-310J, "or by the constant current electrometer "Takeda Riken-84M." The output signal of either device was fed to the input of an automatic recorder or storage oscilloscope. The operational amplifier at two different input impedances had amplification coefficients of 10^{11} V/A and 10^9 V/A, and corresponding time constants of 0.1 sec and <0.01 sec. The output noise of the amplifier did not exceed 100 μV.

The limits of measurements of the electrometer and the corresponding time constants (the time of establishment of 99% of the output signal) at the input impedances 10^8, 10^{10}, and 10^{12} ohms amounted to $3\times10^{-7}-10^{-11}$ A, $3\times10^{-9}-10^{-13}$ A, and $3\times10^{-11}-10^{-15}$ A at <0.7 sec, 1 sec, and 10 sec, respectively.

Figures 9c and d show the experimental dependences of the collector current on the time for two different crystals. A pulse of capacitive current is observed on the experimental curves at the initial instant of time. This pulse characterizes the response of the measurement system to a change in the voltage U_1, the duration of which, depending on the circuit parameters, varied from several milliseconds to several seconds. Taking into account the finite time of passage of the front of the charges between the source and the grid, and selecting one of the five possible measurement regimes, we succeeded in all cases in obtaining a reliable resolution of the capacitive spike and the signal due to the passage of charges through the drift space.

The theoretical dependence of the collector current on the time (Fig. 9b) evidently takes place upon satisfaction of the following conditions:

1) parallelism of the electrodes of the measurement system;

2) homogeneity of the field in the drift space;

3) constancy of the drift velocity of the charges in the given electric field.

While the first two conditions impose definite requirements on the construction of the measurement system, the last condition can be satisfied only in the presence of a sufficiently homogeneous medium filling the entire volume of the triode.

A small disorientation of the electrodes should first of all lead to a decrease in the derivative on the curve of

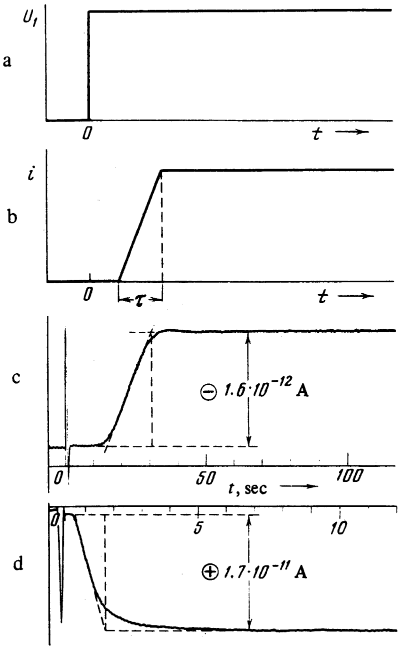

FIG. 9. Measurement of the drift times of the charges: a) time dependence of the voltage between the source and the grid; b) theoretical line dependence of the collector current of the triode strip-chart record of the collector current in solid helium: c) negative charges, $E=20$ kV/cm, $p=25.8$ atm, $T=1.20$ K; d) positive charges, $E=15$ kV/cm, $p=32.4$ atm, $T=1.58$ K.

the time dependence of the current at the beginning and at the end of the linear portion. In the triodes used by us, the roughnesses of the electrodes and the maximum difference in the distances between each pair of electrodes (measured at diametrically opposite points) did not exceed 10 μm in the case of a total value of the drift space of 280 and 300 μm.

The nonuniformity of the field, in addition to the nonparallelism of the electrons and edge effects, can be due to the penetration through the grid into the drift space of the field E_1 and, in addition, to the screening action of the space charge. The absence of an effect of geometric factors on the shape of the experimental curves can be deduced from the numerous records of the collector current obtained in different crystals at sufficiently small space-charge densities, where the time dependence of the current did not actually differ from the theoretical. It is clear that the condition of low space-charge density is equivalent to the condition

$$4\pi ned^2 \ll U_2,$$

where n is the mean charge density in the drift space, and d is the size of the drift gap.

Usually the charge desnity in our experiments amounted to 10^9-10^{10} cm^{-3} at an external field strength of $10^3-5\times10^4$ V/cm. In such a case the space-charge field amounted as a rule to no more than several percent of the value of the external field and had practically no effect on the results of the measurements. Only at maximum densities, $n \gtrsim 10^{11}$ cm^{-3}, was significant distortion of the shape of the collector current observed; there was also a 10–20% change in the time of flight. It should be noted that in some samples the dependence of the collector current on the time differed significantly from the theoretical, while all the conditions enumerated above were satisfied to a sufficient degree. In these cases, the distortions of the shape of the signal were evidently due to the inhomogeneities of the crystal, which affected the value of the drift velocity.

The drift velocity of the charges was determined from the relation $v = d/\tau$. The field strength in the drift space was taken to be equal to $E = U_2/d$. The mobility was calculated in the region of "weak" fields, where the velocity was proportional to the field strength:

$$\mu = v/E = d^2/\tau U_2.$$

In those cases when the observed time dependence of the collector current deviated significantly from the theory, the time τ was determined by extrapolation of the linear portions of the experimental curve to their intercepts (Fig. 9d). Under these conditions, the principal error in the calculation of the velocity and the mobility was determined by the errors in measurement of τ (associated mainly with inaccuracy in the extrapolation) which, depending on the shape of the observed signal, varied from several percent to several dozen percent. The measurements were carried out in the range of fields $10^3-5\times10^4$ V/cm. The times of flight were varied from 20 μsec to 600 sec.

Lacking the possibility of directly monitoring the "quality" of the grown samples, we were forced to judge them, as before, only on the basis of the change in the induced electrical conductivity in the transition from the liquid to the solid phase. However, this criterion is extremely arbitrary for several reasons. First, as is seen from the diode volt-ampere characteristics, the ratio of the currents before and after solidification generally depends, for a given crystal, on the applied voltage and the intensity of the ionization source, and for different diodes this ratio also depends on L. Only in the limit as $U \to 0$ does the ratio of the currents become theoretically proportional to the ratio of the mobilities of the carriers in the liquid and solid helium. Furthermore, the problem of the dependence of the mobility on the quality of the crystal (monocrystallinity, concentration of point defects and dislocations) and on its orientation was not taken into account in sufficient measure, either theoretically or experimentally. Under these conditions, principal significance attaches to the reproducibility of the results of investigations of different samples and to the absence of an "annealing" effect.

From the point of view of reproducibility of the results, all our crystals could be divided into two categories: 1) crystals grown from He I; 2) crystals grown from He II. Figure 10,I shows the results of measurements of the mobility of negative ions in solid He^4 at a pressure of 32.4 atm for thirteen different samples; nine samples were grown in a glass container (Fig. 1A), the remaining four in a metallic container (Fig. 1B). All the crystals were annealed before measurement at a temperature 0.1–0.2 K lower than the melting temperature for periods of 2–3 h. Measurements were carried out on one of the crystals both before and after twelve hours of annealing. In all cases, an exponential depen-

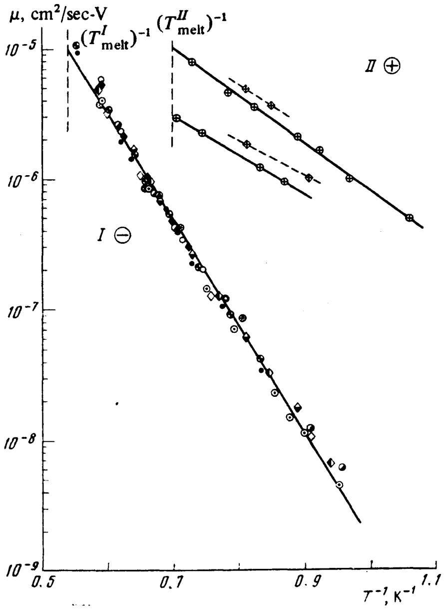

FIG. 10. Temperature dependence of the mobility of negative charges for thirteen samples at a pressure of 32.4 atm (I) and positive charges for four samples at a pressure of 25.8 atm (II). The circles and squares refer respectively to crystals grown in glass or metallic containers.

dence of the mobility on the reciprocal temperature was observed, with an activation energy of $\varepsilon = 19 \pm 1$ K. The values of the mobility at a certain temperature varied for the different crystals by not more than 50%.

Much poorer reproducibility of the results was observed for crystals grown from He II. In the absence of a temperature gradient, it was rather difficult to obtain a uniform growth of the crystal at a specified rate, and this affected directly the results of the measurements. The results shown in Fig. 10, II, which refer to four different samples grown at a pressure 25.8 atm, differ in absolute value by a factor of about four, while activation energy varies from 6.4 to 8.6 K.

Using the time-of-flight method, we measured the dependence of the drift velocity of the ions on the electric field strength and the temperature dependence of the mobility in He^4 crystals at the following pressures and corresponding molar volumes: 50 atm (19.60 cm^3), 40 atm (20.07 cm^3), 36.6 atm (20.27 cm^3), 32.4 atm (20.47 cm^3), 28.1 atm (20.76 cm^3), 25.8 atm (20.93 cm^3). The character of the change in the drift velocity with increase in the field intensity for ions of both signs at all pressures (with the exception of positive ions at $p = 25.8$ atm) can be traced by using as an example the results obtained by us for positive ions in a crystal at a pressure of 32.4 atm (Fig. 11). At a temperature close to the melting temperature ($T = 1.70$ K), in fields up to 2×10^4 V/cm, the velocity is proportional to the field intensity. As the temperature is decreased, the region of linear dependence $v(E)$ becomes narrower. In "strong" fields, a faster than linear growth in the velocity is observed. Possibilities of measurements of the velocity in "weak" fields at low temperatures were limited by the smallness of the measured currents and by the large drift times.

In some experiments, measurements of the velocity were carried out in fields up to 5×10^4 V/cm. Figure 12 shows the dependence of the velocity on the field strength for negative ions at a pressure of 36.6 atm and temperature 1.43 K in the coordinates $v(E)$ and $v^{1/3}(E)$. As is seen from the figure, in the given case, $v \propto E$ in fields not exceeding 10^4 V/cm (curve 1). In fields $>10^4$ V/cm the relation $v = \alpha(E + E_0)^3$ is valid with high accuracy (curve 2). The parameter E_0, determined by extrapolation of the linear part of the $v^{1/3}(E)$ dependence to the intercept with the abscissa, is equal to 1.7×10^4 V/cm.

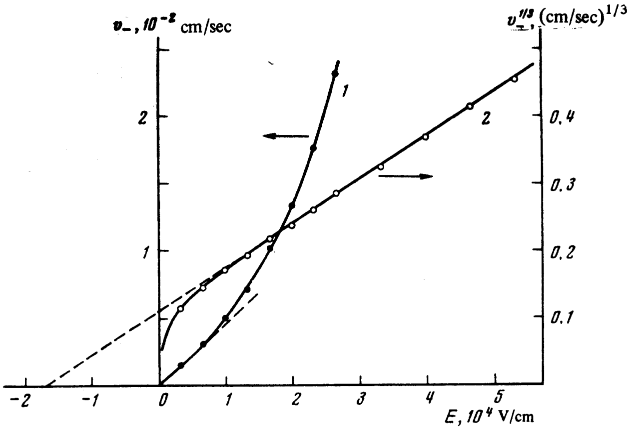

FIG. 12. Dependence of $v_-(E)$ (curve 1) and $v_-^{1/3}(E)$ (curve 2) at $T = 1.43$ K, $p = 36.6$ atm.

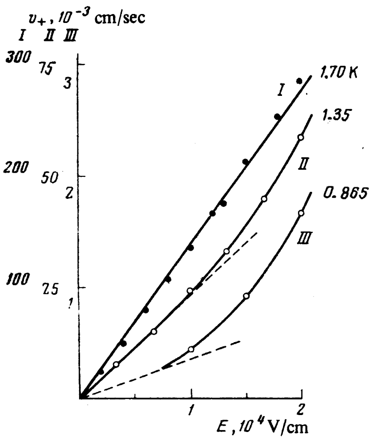

FIG. 11. Dependence of the drift velocity of positive ions on the field strength at different temperatures and a pressure of 32.4 atm. The scale on the ordinate is different for all three curves.

The data considered in Fig. 11 for the positive ions at a pressure of 32.4 atm are represented in the coordinates $v^{1/3}(E)$ in Fig. 13. The characteristic feature is the decrease of E_0 with decrease in temperature.

As has already been noted, for positive ions at a pressure of 25.8 atm, a completely different dependence of the velocity on the field strength was observed (Fig. 14). The $v(E)$ dependences measured in this same crystal at different temperatures for negative ions do not differ qualitatively from the results considered above at higher pressures.

To obtain the temperature dependence of the mobility over a wide range of temperatures, it was necessary to carry out the measurements at a sufficiently large value of the field strength which, however, should be "weak" in the sense of a linear dependence of the velocity. The results set forth were obtained in a field of 10^4 V/cm, which satisfied both requirements in the best fashion. Only in the case of positive ions at 25.8 atm did the temperature dependence of the mobility correspond to the field strength 5×10^3 V/cm.

Figure 15a shows the temperature dependences of the mobility of the positive ions in He^4 crystals at six dif-

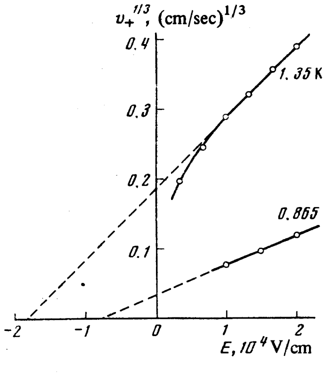

FIG. 13. Dependence of $v_+^{1/3}(E)$ at two different temperatures; $p = 32.4$ atm.

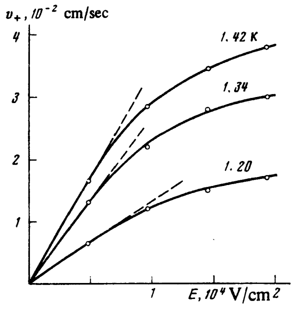

FIG. 14. Dependence of the drift velocity of positive ions on the field strength at different temperatures; $p = 25.8$ atm.

ferent values of the pressure. At all pressures, over a wide temperature range beginning with the melting temperature, an exponential dependence of the mobility on the reciprocal temperature existed:

$$\mu_+ = \mu_0 \exp(-\varepsilon_+/T).$$

The activation energy ε_+, determined from the slope of the linear portion of the plot of $\log\mu_+(1/T)$, increases with increase in the pressure.

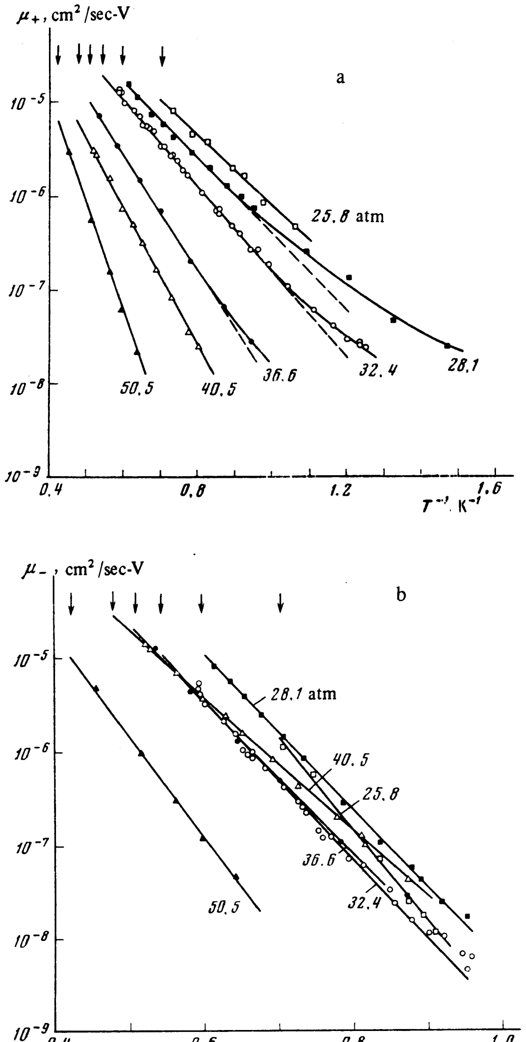

FIG. 15. Temperature dependence of the mobility of positive (a) and negative (b) ions in hcp He^4 at different pressures. The arrows indicate the melting temperature corresponding to these pressures.

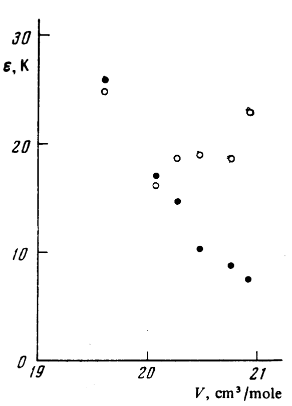

FIG. 16. Dependence of the activation energy on the molar volume for positive (●) and negative (○) ions.

The systematic deviation of the dependence of the mobility from exponential, observed at low temperatures, is evidently due to the fact that at these temperatures fields ~10^4 V/cm are no longer "weak," which leads to a dependence of the mobility on the field strength. At a pressure of 32.4 atm and a temperature 0.865 K ($T^{-1} = 1.16$ K^{-1}), this can be seen from the corresponding curve of Fig. 11. Moreover, the departures of the dependence $\mu(T)$ from exponential are greater the broader the temperature interval, and can be connected with the temperature dependence of the factor μ_0 in front of the exponential; the form of this factor cannot be determined within our limits of our accuracy. If $\mu_0 \propto T^n$, the activation energy obtained by the method shown above exceeds the actual value of ε by an amount ~nT.

The mobility of the negative ions as a function of the reciprocal temperature, for crystals of different density, is shown in Fig. 15b. The results shown in Fig. 15 at the corresponding pressures refer to the same samples.

The dependences of the activation energies ε_+ and ε_- for positive and negative charges on the molar volume in the hcp phase are shown in Fig. 16.

The results of measurements of the mobility of ions of both signs in the bcc phase of He^4 at a pressure of 28.1 atm agree qualitatively with the data obtained earlier by Marty and Williams.[10] The mobility of negative ions in bcc He^4 is 50 times greater than in the hcp phase near the phase transition. The mobility of positive ions in the bcc phase is approximately one half its value in the hcp phase. The measurements of the temperature dependence of the mobility in the bcc phase were not carried out because this phase exists in a small temperature range.

Three successive stages of growth of the crystal are displayed on the photographs (Fig. 17) obtained in the process of crystallization of one of the samples at a pressure of 28.1 atm in a glass container. Initially (Fig. 17a) a large part of the volume of the container, together with the electrodes, was filled with He II. In the lower part, around the copper cold finger, a layer of bcc phase was formed. The shape of the phase boundary is determined by the shape of the isotherms in the

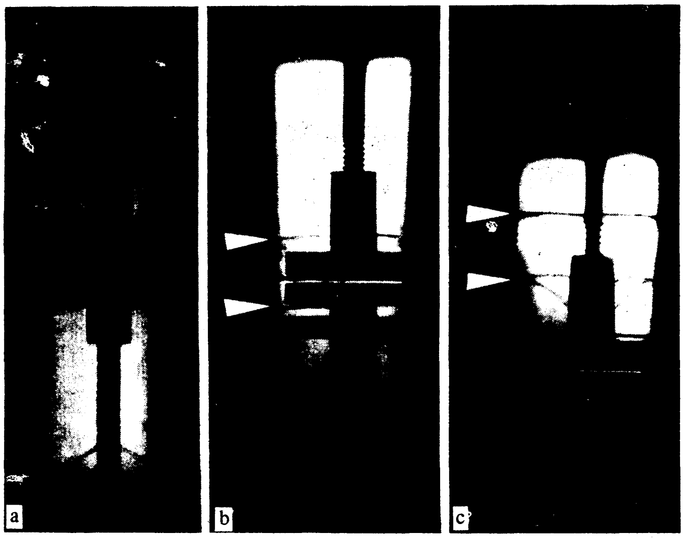

FIG. 17. See explanation in text.

lower part of the container. Next (Fig. 17b), the interelectrode space was filled with the bcc phase. The liquid phase was situated above it and the solid hcp phase below. Both phase boundaries are marked by arrows. The end of the crystallization is observed in Fig. 17c. The liquid (He II) and solid bcc and hcp phases are arranged successively from top to bottom. The interelectrode space was filled with the hcp phase. The dimension of the bcc phase along the axis of the container corresponds to a temperature gradient of 15 mK/mm. The conical shape of the phase boundaries on the photographs of Figs. 17b and c is a consequence of the slight (~ 20 mK) heating of the electrodes due to illumination of the container through the open window of the metallic cryostat by the external light source.

DISCUSSION OF RESULTS

The study of diffusion processes in crystalline He^4 follows two directions at the present time. One is the study of the mobility of carriers of both signs, induced in the helium crystals; the other is the study of the diffusion of the impurity atoms of He^3 by NMR methods. In spite of the difference in structure of all three studied objects, a number of common features have been observed in their diffusion.

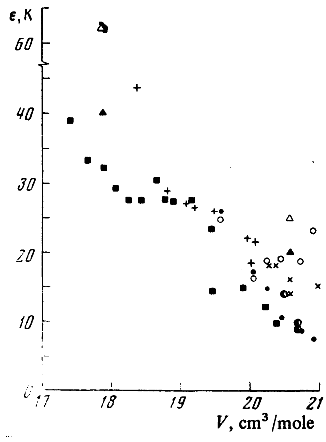

FIG. 18. Dependence of the activation energy of diffusion of positive and negative ions and impurity atoms of He^3 in hcp He^4 on the molar volume. Positive ions: ●—results of present research, ▲—Refs. 9, 12, 13, ■—Ref. 10; negative ions: ○—results of present research, △—Refs. 9, 12, 13; He^3 atoms: ◐—Ref. 17, ×—Ref. 16, +—Ref. 18.

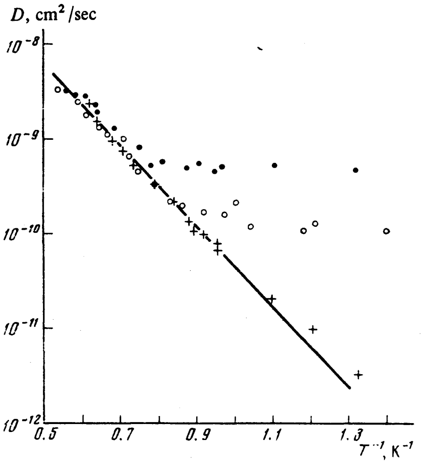

FIG. 19. Temperature dependence of the diffusion coefficient of positive ion (+, results of present research, $V=20.76$ cm³ mole and He^3 atoms ($V=20.7$ cm³/mole, ●—0.75% He^3, ○—2.17% He^3,[17] in hcp He^4. The straight line is described by the equation $D=6.6\times10^{-7}\exp(-9.5/T)$.

1. In accord with the data obtained earlier[9,10,12,13,16–18] and the results of the present work, the diffusion of ions of both signs and of the impurity atoms has an activation character over a rather wide temperature range below the melting temperature. Figure 18 shows the dependence of the activation energies3) ε_+, ε_-, and ε_3 on the molar volume, obtained in the studies listed at the beginning of this paragraph. At molar volumes >18.8 cm³, the activation energies ε_+ and ε_3 are virtually identical. ε_- takes on approximately the same values as ε_+ and ε_3 in the range of molar volumes $19.5\ \text{cm}^3 < V < 20.1\ \text{cm}^3$.

2. The diffusion coefficients $D_\pm = \mu kT/e$ (Fig. 15) and D_3 (see Ref. 17) are practically equal (~ 10^{-9} cm²/sec) and do not depend on the molar volume along the melting curve. It is interesting to note that the temperature dependence of $D_+(T)$ obtained in the present work for $V=20.76$ cm³/mole, and the temperature dependence of $D_3(T)$ measured in Ref. 17 at $V=20.7$ cm³/mole coincide, within the limits of error, over the entire interval of existence of the activation dependence of D_3 (Fig. 19).

3. In strong electric fields, the character of the change in the drift velocity with increase in the field is the same for ions of both signs.

The enumerated experimental data allow us to assume that there exists a single transport mechanism for the ions and impurity atoms.

The problem of the motion of negative ions in solid helium was first considered by Shikin[11] under the assumption that their transport is effected by diffusion of the vacancies that arise around the negative ion under the action of the external electric field. The indicated mechanism explains qualitatively the observed exponential temperature dependence of the mobility and gives the correct order-of-magnitude value of the mobility of the negative ions; however, it is not suitable for the description of the diffusion of positive ions. Of great importance in this case would be a comparison of the theoretical and experimental results in the region of strong electric fields, where a nonlinear dependence is observed in the drift velocity of the ions as a function of the field strength. Unfortunately, it is not possible to carry out such a comparison at the present time in view of the absence of theoretical predictions at sufficiently strong fields.

As was shown by Andreev and Meĭerovich,[19] the motion of the ions at not too low temperatures can be due to inelastic scattering of longwave vacancions from them, accompanied by a transition of the ions to the neighboring crystal-lattice sites. The drift velocity of the ions under these conditions is expressed in terms of the inelastic scattering cross section σ and the parameter of the vacancion energy spectrum and does not depend on the specific structure of the ions. In this connection, the final results obtained in Ref. 19 under the assumption $\Delta \gg T$ (Δ is the vacancion bandwidth) have a common character for ions and impurity atoms; in particular, the temperature dependence of the mobility is determined by the relation

$$\mu(T)=\mu_0 T^{1/2} \exp(-\varepsilon_0/T)$$

(ε_0 is the energy corresponding to the bottom of the vacancion band). The coefficient μ_0, which depends on the interatomic separation a and the bandwidth Δ, is a function of the molar volume only.

Thus, the considered theory explains qualitatively the observed agreement of the diffusion activation energy of the ions and the impurity atoms. However, the quantitative comparison of the results, shown in Fig. 19, and the theory leads to too great a value $\Delta \sim 100$ K, while the actual bandwidth probably does not exceed several degrees. This result can evidently be explained by the fact that in actuality the inelastic-scattering cross section is much less than the value $\sigma = a\hbar/p$ chosen in Ref. 19 in accordance with the condition $U(r)|_{r\approx a} \leqslant \Delta$ ($U(r)$ is the potential of the vacancion–ion interaction). Since data on the value of Δ are lacking, we can only estimate σ, assuming that $\Delta \sim T$ and $D \sim a^2 nu\sigma$ (n is the vacancy concentration and u is their velocity). Setting $n \sim a^{-3} \times \exp(-\varepsilon_0/T)$ and $u \sim a\Delta/\hbar$, we obtain

$$D \sim \frac{\sigma\Delta}{\hbar} \exp\left(-\frac{\varepsilon_0}{T}\right) = D_0 \exp\left(-\frac{\varepsilon_0}{T}\right).$$

For the case considered in Fig. 19, $D_0 = 6.6\times10^{-7}$ cm^2/sec, $T \sim 1$ K and $\sigma \sim 5\times10^{-18}$ cm$^2 \approx 4\times10^{-3} a^2$. The inelastic scattering cross section can be so small for the following reason:

The interaction radius R of a vacancion with an ion or an impurity atom is determined by the relation $U(r) \sim \Delta$, since the kinetic energy of the vacancion cannot change by more than Δ. In the case in which $U(a) \gg \Delta$, the condition $R \gg a$ is satisfied, and here the probability of penetration of the vacancion into the region $r \ll R$ is very small. The inelastic scattering of interest to us is possible only upon penetration of the vacancion into the lattice site adjacent to the ion. Thus, at $U(a) \gg \Delta$, the inelastic scattering cross section is $\sigma \ll a^2$, while the total cross section is much greater than a^2. What is incomprehensible, however is the numerical agreement of the inelastic cross sections for positive ions and He^3 atoms, since σ under these conditions depends essentially on the form of the potential energy $U(r)$.

The significant difference between ε_+ and ε_- at larger molar volumes is difficult to explain within the framework of the theory advanced in Ref. 19 without invoking some additional considerations concerning the character of the interaction of the vacancies with the negative ions. Thus, for example, in the case of resonance scattering, the possibility of which was discussed by the authors of Ref. 19, ε_- can be determined to a significant degree by the parameters of the resonance cross section.

The dependence of the drift velocity of the ions on the field strength, obtained in Ref. 19 under the assumption $e\mathbf{E}\cdot\mathbf{a} < \Delta$, is of the form

$$v \propto \sum_n \mathbf{a}_n [1-\exp(-e\mathbf{E}\mathbf{a}_n/T)] \qquad (5)$$

($\mathbf{a}_n$ is the vector joining the crystal lattice site with the n-th nearest neighbor, and the summation is carried out over the sites for which $e\mathbf{E}\cdot\mathbf{a} > 0$).

In weak fields ($e\mathbf{E}\cdot\mathbf{a} \ll T$), in accord with (5), $v \propto E$, but in strong fields ($e\mathbf{E}\cdot\mathbf{a} > T$) the velocity is practically independent of E. Thus, the character of the $v(E)$ dependence changes substantially in fields $E \sim T/ea$, which corresponds at $T = 1.5$ K to a field strength $\sim 4\times10^3$ V/cm. When formula (5) is compared with the results shown in Fig. 14, attention can be called to a certain qualitative similarity between the results of the theory and experiment.

It is obvious that the considered mechanism of motion of the ions is impossible in fields $e\mathbf{E}\cdot\mathbf{a} > \Delta$, since the vacancy energy cannot change by an amount exceeding Δ. In such strong fields, however, the transport of ions by scattering of vacancions from them, accompanied by the spontaneous emission of phonons, is possible. According to Refs. 19 and 20, the probability of spontaneous emission of phonons is proportional to the cube of the frequency ω and, since $\hbar\omega = e\mathbf{E}\cdot\mathbf{a}$, we have in this case $v \propto E^3$.

The results that we obtained for positive and negative ions (Figs. 12 and 13) are described with good accuracy by the relation

$$v \propto (E+E_0)^3. \qquad (6)$$

It should be noted that the theoretical dependence $v \propto E^3$ was obtained without account of the difference in the energies of the initial and final states of the ion vacancy ε_1 and ε_2. More accurately, the frequency of the phonon should satisfy the relation

$$\varepsilon_1 + e\mathbf{E}\mathbf{a} = \varepsilon_2 + \hbar\omega.$$

As was shown by Meĭerovich, the dependence of the ion velocity on the field strength is in this case of the form

$$v(E)=\mathrm{const}\cdot\int u\sigma\exp\left(-\frac{\varepsilon}{T}\right)d^3u\int\limits_{\omega_1}^{\omega_2}\omega^3\,d\omega,$$
$$\hbar\omega_1=\varepsilon+eEa-\Delta,\quad \hbar\omega_2=\varepsilon+eEa.$$

to within terms of order $(\Delta/eEa)^2$ we have

$$\int\limits_{\omega_1}^{\omega_2}\omega^3\,d\omega=\frac{\Delta}{\hbar^4}(eEa)^3\left(1+3\frac{\varepsilon-{}^1/_2\Delta}{eEa}\right)$$

and

$$v(E)=\mathrm{const}\cdot E^3\left[1+3\frac{F(T)-{}^1/_2\Delta}{eEa}\right]\cong\mathrm{const}\cdot\left[E+\frac{F(T)-{}^1/_2\Delta}{ea}\right]^3, \qquad (7)$$

where

$$F(T)=\int\exp\left(-\frac{\varepsilon}{T}\right)\varepsilon u\sigma\,d^3u\Big/\int\exp\left(-\frac{\varepsilon}{T}\right)u\sigma\,d^3u.$$

Since we are talking about strong fields, it is necessary to establish more precisely the degree to which $F(T)$ does not depend on E. According to the results of Ref. 20, the inelastic scattering cross section σ can be assumed to be independent of E in fields satisfying the relation $eEa \ll 10\,\Delta$.

At low temperatures ($T \ll \Delta$), by assuming that $\sigma\propto u^{-1}$, $\varepsilon\propto u^2$, as was done in Ref. 19, we obtain $F(T)=(3/2)T$ and

$$v(E)=\mathrm{const}\cdot\left(E+\frac{3T-\Delta}{2ea}\right)^3. \qquad (8)$$

It is easy to see that the formula (7) agrees qualitatively with (6) and describes correctly the character of the change of E_0 with decrease in the temperature. However, it should be noted that the requirement $eEa \ll 10\,\Delta$ in our case ($E_{\max}=5.3\times10^4$ V/cm) corresponds to the inequality $\Delta \gg 2$ K. On the other hand, the change in the character of the dependence $v(E)$ observed at a field strength $\sim 10^4$ V/cm, in the case when this change is due to the emission of phonons, should take place in fields $eEa\sim\Delta$, whence it follows that $\Delta\sim 4$ K. As a result, the last two estimates of the bandwidth agree only qualitatively with each other.

Thus, the results do not contradict the considered theory; however, for its final confirmation, more detailed experimental data are necessary. It is interesting to note that if the vacancion mechanism of ionic, motion is valid the results of the measurements of the dependence of the velocity in strong fields at sufficiently low temperatures $T \ll \Delta$, as follows from formula (8), make it possible to determine the vacancion bandwidth directly.

In conclusion, we take this opportunity to thank P. L. Kapitza for interest in the work, A. I. Shal'nikov for constant attention and great help in completing the research, A. F. Andreev and A. E. Meĭerovich for numerous useful suggestions and discussions.

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[1] The term "ions" used at the present time for carriers of positive and negative charges in solid helium is very tentative and does not correspond to the actual structure of the carriers, especially in the case of negative charges. The structure of the carriers in solid helium has been discussed in detail in Refs. 9–11.

[2] The value of the current at $U_1=0$ was taken as its zero value in practice.

[3] The quantities denoted by the indices +, –, and 3 refer respectively to positive and negative ions and to He^3 atoms.

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