Ion Motion in Superfluid Liquid Helium under Pressure*

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Recent investigations of superfluidity by a study of the mobilities of ions in liquid He II have been extended to the liquid under pressure. At a fixed temperature the positive-ion mobility decreases appreciably as the pressure is increased, particularly at low temperatures. At a fixed pressure the mobility increases less rapidly with decreasing temperature at higher pressures. The negative-ion mobility, smaller than that of the positive ion at zero pressure, becomes equal to that of the latter above 7 atm. In high electric fields and at high pressures, the drift velocity of the negative ions approaches a limiting value roughly equal to the Landau critical velocity for a body moving through the superfluid. The theory, which discusses the mobility in terms of ion scattering by rotons and phonons, is reviewed. It is pointed out that previously neglected effects concerned with the importance of small-angle scattering of the ion ought to be taken into account; some earlier estimates of scattering cross sections are revised accordingly. It is then shown that this theory, making use only of the known change of the roton dispersion relation with pressure, can account quantitatively for the observed pressure dependence of the positive-ion mobilities.

I. INTRODUCTION

We have recently reported[1–3] experiments designed to investigate the superfluidity of liquid helium on a microscopic scale by a study of the motion of ions immersed in the fluid. The ions were produced by α particles from a Po^{210} source and their drift velocity u in the presence of an applied electric field $\mathcal{E}$ was measured directly by a time-of-flight method. The temperature dependence of the mobility $\mu=u/\mathcal{E}$ could be interpreted in terms of the scattering of the ions by the various elementary excitations (quasi-particles) which characterize the quantum fluid. In order to subject this interpretation and the quasi-particle description to a further check, it seemed of interest to extend our measurements to liquid helium under pressure. In going to 25 atm, the melting pressure of liquid helium at 0°K, the density of the liquid increases by about 18%, and the dispersion relation of the elementary excitations is appreciably affected. As a result, significant changes of ion mobilities can be expected as the pressure on the liquid is increased. We here report the results of such measurements.

II. EXPERIMENTAL ASPECTS AND RESULTS

The apparatus described in reference 2, Fig. 2, was modified in the following way. The $\frac{3}{16}$-in. tube, which had been used to fill the experimental chamber with helium from the small gas holder, was replaced by a stainless steel capillary of 1 mm i.d. This capillary was then used to connect the experimental chamber with a high-pressure He cylinder via a charcoal trap at liquid-nitrogen temperature. The pressure in the system could be measured by a Bourdon gauge with an accuracy of ±1%. The fact that under these circumstances a filament of liquid helium in the different capillaries connected the experimental chamber with warmer parts of the cryostat resulted in an increased heat leak into the apparatus. The lowest temperature which could readily be reached with the He^3 refrigerator was consequently only about 0.51°K and the temperature stability was also somewhat impaired. The electronic circuitry was the same as described in reference 2.

Measurements above 1°K were performed with the vacuum jacket removed and the experimental chamber in direct thermal contact with the main helium bath. Under these conditions the ion mobility could be measured as a function of pressure at constant temperature because the large heat capacity of the bath absorbed easily the thermal effects of pressure changes. Below 1°K, however, changes in pressure produced pronounced changes in temperature so that it appeared preferable to make measurements at different temperatures for fixed values of the pressure.

At each temperature and pressure the mobility of positive and negative ions was measured in immediate succession simply by reversing the polarity of the grid potentials.

The experimental results for the mobility $\mu=u/\mathcal{E}$ obtained in the limit of sufficiently small field $\mathcal{E}$ are presented in Figs. 1 and 2.[4] The temperature dependence of the mobility for various values of the pressure is shown in Fig. 1 which is a plot of $\log_{10}\mu$ vs T^{-1} of the same kind as used in our previous work.[2,3] The points below 1°K were measured directly; those above 1°K were taken from the curves of Fig. 2 for the corresponding pressures. Figure 1 shows that, at each pressure, $\ln\mu$ is a linear function of T^{-1}. The slope of these lines decreases with increasing pressure, being for positive ions equal to 8.8°K at 0 atm, and to 7.6°K at 21.4 atm.

Figure 2 shows a plot of $\log_{10}\mu$ as a function of pres-

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[1] L. Meyer and F. Reif, Phys. Rev. **110**, 279 (1958).
[2] F. Reif and L. Meyer, Phys. Rev. **119**, 1164 (1960).
[3] L. Meyer and F. Reif, Phys. Rev. Letters **5**, 1 (1960).

[4] Several runs at slightly different pressures yielded results consistent with those of Fig. 1 but were omitted from the graph to avoid confusion.

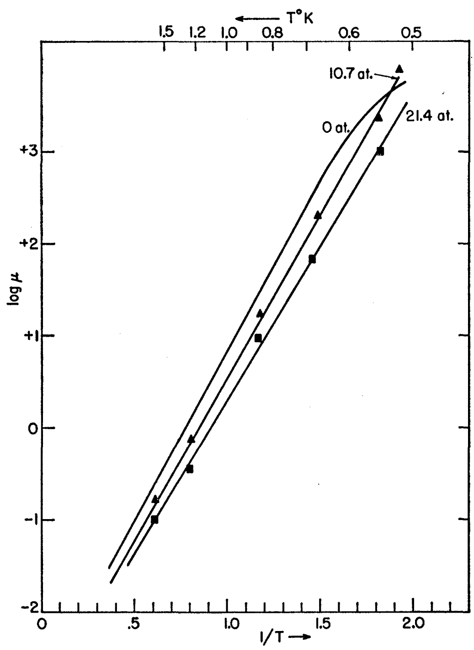

FIG. 1. Temperature dependence of the positive-ion mobility at various pressures. The mobility μ is measured in $cm^2\ v^{-1}\ sec^{-1}$; the temperature T is in deg K. At 10.7 and 21.4 atm the mobility of the negative ion is equal to that of the positive one.

sure at various temperatures. The curves for $T=1.23$°K and $T=1.64$°K were measured directly; the others are derived from cuts at constant T through the curves of Fig. 1. The zero-pressure values are taken from Fig. 1 of reference 3. It is seen that for positive ions $\ln\mu$ is a linearly decreasing function of the pressure. For example, increasing the pressure from 0 to 25 atm at 1.23°K decreases the mobility by a factor of 5; at 0.57°K the decrease is approximately by a factor of 14. Recently Cunsolo and Mazzoldi[5] have also made measurements of ion mobilities under pressure for temperatures above 1.1°K. Their results for positive ions are in agreement with ours in the temperature range where they overlap.

At zero pressure, the curves of $\ln\mu$ vs T^{-1} fall below the straight-line relationship at sufficient low temperatures[3] (below about 0.7°K for positive ions). This behavior has been attributed[3] to the emergence of another ion scattering mechanism (phonon scattering) at low temperatures. It is to be noted, however, that at higher pressures the curves of Fig. 1 remain straight down to 0.51°K, the lowest temperature investigated. Furthermore, extrapolation at low temperatures of curves of $\ln\mu$ vs pressure $\mathcal{P}$ to zero pressure yields values of μ which are not those actually measured at $\mathcal{P}=0$, but which instead lie on the extrapolated straight line of $\ln\mu$ vs T^{-1} for $\mathcal{P}=0$. These results indicate that at higher pressures the other scattering mechanism due to phonons does not become important down to the lowest temperatures investigated.

The behavior of the negative-ion mobility is remarkable and somewhat surprising. At zero pressure the mobility of the negative ion is always less than that of the positive one[3]—at low temperatures by more than a factor of 10. On the other hand, one finds that, irrespective of the temperature, the negative-ion mobility becomes equal to that of the positive ion (to within the experimental error of $\pm 2\%$) at pressures above about 7 atm. As the pressure is increased at a fixed temperature, the negative-ion mobility thus increases initially so as to approach the larger positive-ion mobility, and then decreases like the latter as the pressure is increased further.

Finally we should like to mention some unexpected results concerning the drift velocity of ions in large electric fields. When the applied field $\mathcal{E}$ is sufficiently large for the ion to acquire energy in excess of thermal energy, the drift velocity is no longer proportional to $\mathcal{E}$. It has also been pointed out[2] that when $\mathcal{E}$ is made large enough an ion can lose momentum and energy by *creating* a roton in the liquid, and that its drift velocity should then tend to approach a limiting value roughly equal to the Landau critical velocity[6] for a body moving through the superfluid, i.e., of the order of $\Delta/p_0=60$ m/sec at zero pressure (or 45 m/sec at 25 atm).[7] In actual experiments it proved, however, impossible to make measurements in the high-field region because of the failure of the gating grids to control the ion

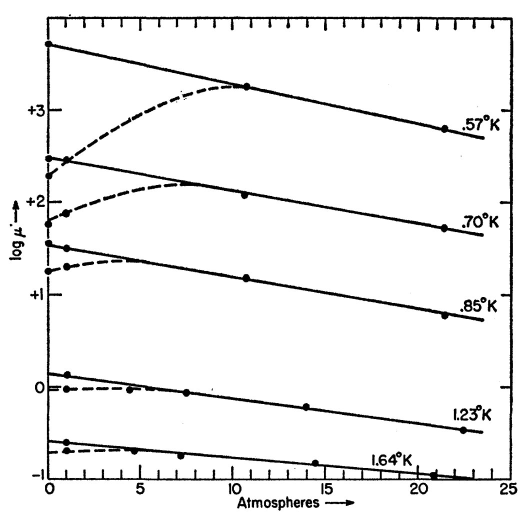

FIG. 2. Pressure dependence of the mobilities at various temperatures. The mobility μ is measured in $cm^2\ v^{-1}\ sec^{-1}$, the pressure $\mathcal{P}$ is in atm. The solid curves refer to the positive ion, the dashed curves to the negative ion. The point at 0.57°K and zero pressure was obtained by extrapolating the curve of $\ln\mu$ vs T^{-1} to this temperature, neglecting its bending over due to phonon scattering.

[5] S. Cunsolo and P. Mazzoldi (unpublished). We wish to thank Professor G. Careri for sending us some of these unpublished data.

[6] See, for example, E. M. Lifshitz, *A Supplement to "Helium"* (Consultants Bureau, New York, 1959).

[7] The symbols and numerical values are those used in the discussion section.

current adequately and consequent loss of resolution in our time-of-flight method. In some experiments with dc potentials, this difficulty was traced to a peculiar "runaway" behavior of the ions in high fields.[8] This behavior is characterized by the tendency of the ions under these circumstances to behave almost like free particles which cannot be stopped simply by a retarding field, but only by a retarding potential greater than the accelerating potential. Although this runaway phenomenon is exhibited by positive ions at all pressures and by negative ions at low pressures, we found to our surprise that it does not seem to occur for negative ions at high pressures, where good measurements of the drift velocity are then possible. There appears to be a limiting drift velocity which is, at 0.55°K and 23 atm, about 52 m/sec; and at 0.51°K and 15 atm, about 59 m/sec. The experimental data are shown in Fig. 3.

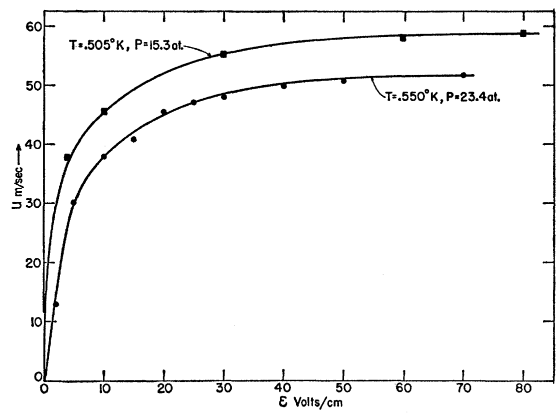

FIG. 3. Dependence of the drift velocity of the negative ion as a function of electric field at low temperatures and high pressures. Limiting drift velocity at 0.550°K and 23.4 atm about 52 m/sec and at 0.505°K and 15.3 atm about 59 m/sec.

III. DISCUSSION

A. General Considerations

In previous work[1–3] we interpreted the temperature dependence of the ion mobilities in terms of the scattering of ions by the elementary excitations (quasiparticles) of the quantum fluid. We now wish to review this interpretation and to examine the results expected on this basis if the density of the fluid is increased by the application of pressure.

Considering an ion of charge e and effective mass M in an electric field $\boldsymbol{\mathcal{E}}$, the expression for the mobility μ can be obtained by equating $e\mathcal{E}$, the gain per second of ion momentum from the field, to $\langle P\rangle_c$, the mean loss per second of ion momentum along the direction of $\boldsymbol{\mathcal{E}}$ due to collisions with the excitations of the fluid. Calculation of $\langle P\rangle_c$ involves a properly weighted angular average of the differential scattering cross section. For purposes of a simplified discussion one can estimate $\langle P\rangle_c$ by multiplying τ^{-1}, the probability per second of an ion-excitation collision, by $f(Mu)$, the mean ion momentum loss per collision. Here Mu is the mean ion momentum in the field direction, while f is the mean fraction of its momentum which the ion loses in each collision. Thus one obtains

$$\mu=(u/\mathcal{E})=(e/M)(\tau/f). \tag{1}$$

The factor f was not taken into account in our previous considerations[2,3] (where we put $f=1$) and it deserves some comments. This factor measures the importance of small-angle scattering, i.e., of "persistence of velocity" effects. If large-angle scattering is predominant (scattering of a "light" ion by a relatively "heavy" excitation), then $f\approx 1$; if small-angle scattering is predominant (scattering of a "heavy" ion by a relatively light excitation), then $f\ll 1$. Since the effective mass of the ion is likely to be large, the second situation is of importance. For purposes of comparing theoretical estimates of scattering cross sections with experimental results, such persistence of velocity effects should, therefore, be kept in mind.[8,9]

The collision probability $\tau^{-1}=N\sigma V$, where N is the number of scattering excitations per cm^3, σ the total scattering cross section between ion and excitation, and V the mean relative thermal velocity between these. $V^2=V_i^2+V_e^2$, where V_i and V_e are the thermal velocities of ion and excitation, respectively. Thus Eq. (1) becomes

$$\mu=(e/M)(V\sigma fN)^{-1}. \tag{2}$$

If the excitation has the classical dispersion relation $\epsilon=p^2(2m)^{-1}$ between its energy ϵ and momentum $\mathbf{p}$ (e.g., if it is a He^3 atom in the superfluid), then calculation of $\langle P\rangle_c$ with hard-sphere or δ-function interaction between ion and excitation yields approximately $f=m/(m+M)$. Also $V=(3kT)^{\frac{1}{2}}(M^{-1}+m^{-1})^{\frac{1}{2}}$. Equation (2) then reduces, except for the exact magnitude of the multiplicative constant obtained by more careful calculation of $\langle P\rangle_c$, to the Langevin expression for the mobility of an ion in a gas[10]:

$$\mu=(\tfrac{3}{8})(\pi/2)^{\frac{1}{2}}e(N\sigma)^{-1}(kT)^{-\frac{1}{2}}(M^{-1}+m^{-1})^{\frac{1}{2}}. \tag{3}$$

Note that for $M>m$, the mobility becomes insensitive to the value of the effective mass M assumed for the ion. The physical reason is that, as M becomes large, the electric field is less effective in accelerating the ion, but collisions are also less effective in deflecting the ion; these two effects tend to cancel each other. Taking persistence of velocity effects into account thus leads

[8] F. Reif. *Seventh International Conference on Low-Temperature Physics, Toronto, 1960* (University of Toronto Press, Toronto, Canada, 1960).

[9] Several attempts have recently been made to calculate the magnitude of ion-excitation scattering cross sections. J. De Boer and A. 'T Hooft, *Seventh International Conference on Low-Temperature Physics, Toronto, 1960* (University of Toronto Press, Toronto, Canada, 1960), p. 304; R. Abe and K. Aizu, Bull. Am. Phys. Soc. **6**, 15 (1961); C. G. Kuper (to be published).

[10] P. Langevin, Ann. chim. et phys. **28**, 317, 495 (1903); also L. B. Loeb, *Basic Processes of Gaseous Electronics* (University of California Press, Berkeley, California, 1955), p. 42.

to estimates of the ion-He^3 cross section σ_{i3} larger than those estimated previously[3] and less dependent on the assumed value of M. For an assumed He^3 effective mass[11] $m=2M_{\rm He}$ ($M_{\rm He}$ being the mass of a He^4 atom), the experimental results[3] combined with Eq. (3) lead to estimates $\sigma_{i3}=1.8\times10^{-14}$ cm^2 for positive ions and $\sigma_{i3}=1.2\times10^{-13}$ cm^2 for negative ions if $M\gg M_{\rm He}$. These estimates would be increased by 40% if $M=m$.

We now consider the scattering of ions by rotons whose dispersion relation is

$$\epsilon=\Delta+(2\mu_0)^{-1}(p-p_0)^2.$$

If the ion effective mass $M<10M_{\rm He}$, the ion momentum $P=(3MkT)^{\frac{1}{2}}$ is still small compared to p_0. In this case large-angle scattering of the ion is possible despite the small value of the mass parameter μ_0 ($\mu_0=0.16M_{\rm He}$), and the roton momentum change will also be small of the order of P. If $\mathbf{s}$ denotes a unit vector along the z axis in the direction of the roton momentum before collision, the roton momentum $\mathbf{p}$ will throughout the collision process be of the form $\mathbf{p}=p_0\mathbf{s}+\mathbf{q}$, where $|\mathbf{q}|\ll p_0$. The roton dispersion relation can then be approximated by $\epsilon=\Delta+(2\mu_0)^{-1}q_z^2$. The roton thus behaves analogously to an electron in a solid which is, in an ellipsoidal conduction band, characterized by a small effective mass μ_0 in one direction and very large (infinite) effective masses in the other two principal directions. An estimate of $\langle P\rangle_c$ for a δ-function type interaction then yields the approximate value

$$f=\tfrac{1}{3}[2+\mu_0/(\mu_0+m)]\approx\tfrac{2}{3},$$

since $\mu_0\ll M$. Persistence of velocity effects thus tend to increase our previous estimates[2] of the ion-roton scattering cross section by about 30%.

The number of rotons is given by[2]

$$N=2(2\pi)^{\frac{3}{2}}h^{-3}(\mu_0kT)^{\frac{1}{2}}p_0\exp(-\Delta/kT). \tag{4}$$

The relative ion-roton velocity is

$$V=[(3kT/M)+(kT/\mu_0)]^{\frac{1}{2}}\approx(kT/\mu_0)^{\frac{1}{2}}$$

since $\mu_0\ll M$. Hence we obtain the approximate expression for the ion mobility in the temperature range where roton scattering is predominant:

$$\mu=\tfrac{3}{4}(2\pi)^{\frac{3}{2}}h^3(e/M)(kT\sigma)^{-1}p_0^{-2}\exp(\Delta/kT). \tag{5}$$

Our discussion of the pressure dependence of the mobility will be based on this expression. Note that, because of a cancellation of terms, it does not contain the mass parameter μ_0.

Finally we mention briefly the scattering of ions by phonons whose dispersion relation is simply $\epsilon=cp$, where c is the velocity of sound in liquid He. The number density of phonons is given by[3] $N=(2.4)(4\pi)(kT/hc)^3$ and, since the ion velocity is small compared to c, $V\approx c$ in Eq. (2). The phonon momentum $p\approx kT/c$ is appreciably smaller than the ion momentum $P=(3MkT)^{\frac{1}{2}}$, so that the ion is scattered only through small angles $\varphi\approx p/P$ with a small momentum change $P(1-\cos\varphi)=\frac{1}{2}P\varphi^2$. Hence $f=\frac{1}{2}(p/P)^2\approx kT(6Mc^2)^{-1}\ll1$ and Eq. (2) becomes, for phonon scattering, approximately

$$\mu=0.2(eh^3k^{-4})\sigma^{-1}(c/T)^4. \tag{6}$$

For a temperature-independent scattering cross section σ, Eq. (6) yields[12] $\mu\propto T^{-4}$ compared with the experimental results[3] which gave for positive ions approximately $\mu\propto T^{-3.3}$. Taking into account the small-angle scattering effects contained in f leads to estimates of the ion-phonon cross section σ_{ip} appreciably larger than previously given. The results of reference 3 combined with Eq. (6) yield at 0.55°K the estimates $\sigma_{ip}=1.5\times10^{-13}$ cm^2 for positive ions and $\sigma_{ip}=3.7\times10^{-12}$ cm^2 for negative ions. The dependence of μ on the velocity of sound c is of significance in discussing the pressure dependence of the mobility.

B. Pressure Dependence of the Mobility

Application of pressure to liquid helium increases its density and thus affects the dispersion relation characterizing the elementary excitations of the superfluid. This dispersion relation for the liquid at low temperatures under its vapor pressure (i.e., essentially at zero pressure) has been determined in a very direct way by neutron scattering experiments.[13] Recently Henshaw and Woods[14] have repeated this kind of experiment for liquid helium at a pressure of 25.3 atm. Hence the dispersion relation is now known at these two pressures. The corresponding parameters characterizing the roton branches of these curves are summarized in Table I. Measurements at intermediate pressures are not available. As a first approximation, we shall estimate the parameters in this range by using linear interpolation

TABLE I. Summary of change of roton parameters with the pressure of liquid helium. The data are those of Henshaw and Woods.[14]

$\mathcal{P}$ (atm)	Δ/k (deg K)	$p_0/\hbar$ (A^{-1})	$\mu_0/M_{\rm He}$
0	8.65	1.91	0.16
25.3	7.0	2.05	(0.09)

[11] I. M. Khalatnikov and V. N. Zharkov, Soviet Phys.—JETP **5**, 906 (1957).

[12] This result is analogous to the low-temperature T^{-5} dependence of the electrical conductivity of metals when the resistivity mechanism is electron-phonon scattering. The extra factor T^{-1} compared to Eq. (6) is due to the fact that for the electron case $f\propto T^2$ since the electron momentum P is the temperature-independent Fermi momentum.

[13] H. Palevsky, K. Otnes, and K. E. Larsson, Phys. Rev. **112**, 11 (1959); J. L. Yarnell, G. P. Arnold, P. J. Bendt, and E. C. Kerr, *ibid.* **113**, 1379 (1959); D. G. Henshaw, Phys. Rev. Letters **1**, 127 (1959).

[14] D. G. Henshaw and A. D. B. Woods, *Seventh International Conference on Low-Temperature Physics, Toronto, 1960* (University of Toronto Press, Toronto, Canada, 1960) p. 64.

formulas of the form:

$$\Delta=\Delta^*(1-a\mathcal{P}), \tag{7}$$

$$p_0=p_0^*(1+b\mathcal{P}), \tag{8}$$

where $\mathcal{P}$ is the pressure, the starred quantities are values of the parameters for $\mathcal{P}=0$, and the positive constants a and b are chosen so that Δ and p_0 agree with the measured values for $\mathcal{P}=25.3$ atm. This requires that $a=7.54\times10^{-3}$ atm^{-1} and $b=2.9\times10^{-3}$ atm^{-1}.

Consider first the temperature range where ion-roton scattering is predominant. Application of pressure changes the roton parameters as shown in Table I and thus increases the number density of rotons in accordance with Eq. (4). Hence Eqs. (2) or (5) lead one to expect a corresponding decrease of the ion mobility. If one makes the simplest assumption that the ion effective mass M and the ion-roton cross section σ are unaffected by the density change of the fluid, one is led to the following predictions.

At a fixed pressure, the temperature dependence of the mobility is described by a plot of $\ln\mu$ vs T^{-1} which is a straight line of slope equal to Δ/k.[15] Such a plot for the liquid at a higher pressure ought to yield a straight line of smaller slope in accordance with Eq. (7). This interpolation equation predicts slopes at 10.7 and 21.4 atm smaller than the slope of the zero-pressure curve by 0.7° and 1.4°K, respectively. For comparison, the experimental curves of Fig. 1 at these two pressures yield slopes smaller than that of the zero-pressure curve by 0.7° and 1.2°K, respectively. The agreement is satisfactory.

At a fixed temperature, the pressure dependence of the mobility should, by Eq. (5), be given by the proportionality

$$\mu\alpha p_0^{-2}\exp(\Delta/kT). \tag{9}$$

A knowledge of the pressure dependence of μ_0 is of no importance. Making use of the interpolation formulas (7) and (8), one obtains to good approximation

$$\ln(\mu/\mu^*)=-[a\Delta^*(kT)^{-1}+2b]\mathcal{P}. \tag{10}$$

Hence a plot of $\ln\mu$ vs $\mathcal{P}$ is expected to yield a straight line with a slope predicted by (10). For example, at 1.64°K the slope should be -0.046 atm^{-1}, about 87% of this value arising from the pressure variation of Δ; at 0.57°K the slope should be -0.12 atm^{-1}, with 95% of this value due to the change in Δ. The experimental curves of Fig. 2 yield slopes at these two temperatures of -0.042 and -0.105 atm^{-1}, respectively. The theoretical model thus accounts quite well for the magnitude of the observed decrease of positive-ion mobility with increasing pressure.

The fact that the mobility at higher pressures shows no evidence for phonon scattering down to the lowest temperatures investigated is also in accord with expectation. If μ_r is the mobility limited by roton scattering and μ_p is that limited by phonon scattering, then the observed mobility μ is given by $\mu^{-1}=\mu_r^{-1}+\mu_p^{-1}$. At 0.5°K and zero pressure, $\mu_p^{-1}=5\mu_r^{-1}$ so that phonon scattering predominates. On the other hand, when at this temperature the pressure is increased to 25 atm, Eq. (5) predicts that the roton scattering measured by μ_r^{-1} is increased by a factor of 30. At the same time, since the velocity of sound c increases from 237 to 365 m/sec,[16] Eq. (6) predicts on this account a decrease in phonon scattering μ_p^{-1} by a factor 5.6. The net result is that at 25 atm and 0.5°K one expects that $\mu_p^{-1}=0.03\,\mu_r^{-1}$, so that phonon scattering should become predominant only at significantly lower temperatures.

C. Concluding Remarks

The quasi-particle description of superfluid liquid helium has previously been shown to explain the pronounced temperature dependence of the ion mobilities in terms of the scattering of ions by rotons and phonons. The present work shows that this theory can also account satisfactorily for the essential features of the observed pressure dependence of the mobilities. Less can be said about the structure of the ions themselves. The fact that the pressure dependence of the positive-ion mobility can be accounted for quite well by the changed roton dispersion relation without having to assume a changed ion effective mass or scattering cross section would seem to argue against Atkins' suggestion[17] that the ion is surrounded by a solid helium sphere which becomes large as the pressure of the liquid approaches its melting pressure. As for the negative ion, it is clear that any detailed model of its structure must explain the fact that its low-field mobility becomes identical to that of the positive ion at higher pressures.

ACKNOWLEDGMENTS

We wish to acknowledge some stimulating conversations with Dr. K. Aizu, Dr. C. G. Kuper, and Dr. W. A. Harrison.

[15] We neglect the pre-exponential factor T^{-1} as discussed in reference 2.

[16] K. R. Atkins, *Liquid Helium* (Cambridge University Press, New York, 1959), p. 130.

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