

72-4783

DIONNE, Vincent Edward, 1942-
THE EFFECT OF PRESSURE ON THE ELECTRON MOBILITY
IN SOLID HELIUM.

The University of Arizona, Ph.D., 1971
Physics, solid state

University Microfilms, A XEROX Company, Ann Arbor, Michigan

THIS DISSERTATION HAS BEEN MICROFILMED EXACTLY AS RECEIVED

THE EFFECT OF PRESSURE ON THE ELECTRON MOBILITY
IN SOLID HELIUM

by

Vincent Edward Dionne

A Dissertation Submitted to the Faculty of the
DEPARTMENT OF PHYSICS
In Partial Fulfillment of the Requirements
For the Degree of
DOCTOR OF PHILOSOPHY
In the Graduate College
THE UNIVERSITY OF ARIZONA

1 9 7 1

THE UNIVERSITY OF ARIZONA
GRADUATE COLLEGE

I hereby recommend that this dissertation prepared under my
direction by Vincent Edward Dionne
entitled The Effect of Pressure on the Electron Mobility
in Solid Helium
be accepted as fulfilling the dissertation requirement of the
degree of Doctor of Philosophy

Carl I. Conzuek July 26, 1971
Dissertation Director Date

After inspection of the final copy of the dissertation, the
following members of the Final Examination Committee concur in
its approval and recommend its acceptance:*

<u>Ray M. Emrich</u>	<u>7-26-71</u>
<u>Jin-Wei Shen</u>	<u>7-26-71</u>
<u>Donald R. Huffman</u>	<u>7-26-71</u>
<u>David E. James</u>	<u>7-26-71</u>
<u>KJ Wouchee</u>	<u>8/19/71</u>

*This approval and acceptance is contingent on the candidate's
adequate performance and defense of this dissertation at the
final oral examination. The inclusion of this sheet bound into
the library copy of the dissertation is evidence of satisfactory
performance at the final examination.

PLEASE NOTE:

**Some Pages have indistinct
print. Filmed as received.**

UNIVERSITY MICROFILMS

STATEMENT BY AUTHOR

This dissertation has been submitted in partial fulfillment of requirements for an advanced degree at The University of Arizona and is deposited in the University Library to be made available to borrowers under rules of the Library.

Brief quotations from this dissertation are allowable without special permission, provided that accurate acknowledgment of source is made. Requests for permission for extended quotation from or reproduction of this manuscript in whole or in part may be granted by the head of the major department or the Dean of the Graduate College when in his judgment the proposed use of the material is in the interests of scholarship. In all other instances, however, permission must be obtained from the author.

SIGNED: Vincent E. Dionne

ACKNOWLEDGMENTS

The author wishes to express his appreciation to Professor C. T. Tomizuka for his encouragement and guidance throughout this research, to Dr. R. A. Young for his helpful discussion of the results, and to Mr. Rodney C. Lowell for his maintenance of the high pressure apparatus. This research project was sponsored by the United States Atomic Energy Commission through Contract AT(11-1)-1041. The support of this agency is gratefully acknowledged.

TABLE OF CONTENTS

	Page
LIST OF ILLUSTRATIONS	v
LIST OF TABLES	vi
ABSTRACT	vii
INTRODUCTION	1
EXPERIMENTAL METHOD	4
SAMPLE PURITY	9
PROBE CALIBRATION	10
EXPERIMENTAL RESULTS	14
DISCUSSION	19
APPENDIX A: OPERATION OF THE PROBE	36
REFERENCES	39

LIST OF ILLUSTRATIONS

Figure	Page
1. Schematic Diagram of the Apparatus	5
2. Cross Sectional View of Assembled Probe	7
3. Calibration Results for Grid II	12
4. Typical Raw Data for Liquid Helium	16
5. Raw Data for Dense Helium Gas	18
6. Pressure Dependence of the Wigner-Seitz Calculation of V_0	26
7. $E_e + E_{pV}$ <u>versus</u> R_0 at 6400 atm	27
8. Surface Energy for Several Values of F_s	29

LIST OF TABLES

Table	Page
1. The Electron Mobility in Four Rare-Gas Solids . .	3
2. Apparatus Sensitivity	13

ABSTRACT

The effect of pressure on the mobility of the cavity-localized electron in solid helium has been studied to a pressure of 6660 atm. No delocalized electron state has been detected at this pressure below the melting point of the solid. It is shown that the results are consistent with the presence of electron bubbles at the highest pressures investigated. The nature of possible charge trapping mechanisms that might account for the results is discussed.

INTRODUCTION

The study of properties of excess electrons in liquid¹⁻³ and gaseous^{4,5} helium has received much attention during the past decade. The localized nature of the excess electron has been demonstrated both experimentally and theoretically. Its configuration as a bubble has been well established. With several exceptions relatively little has been investigated in solid helium in which the electron is also localized. Keshishev, Mezhev-Deglin, and Shal'nikov⁶ made preliminary measurements which have established a lower limit for the mobility of electrons in He⁴ crystals. Cohen and Jortner⁷ have extended theoretical considerations made initially for the liquid and gas phases to the problem of excess electrons in solid helium.

Within a broad range of helium densities the excess electron is self-trapped in a cavity whose radius is several times the inter-atomic distance. The cavity is the minimum energy configuration in helium associated with a weakly attractive long-range electron-helium atom polarization potential and a strong short-range electron-atom repulsion.⁸ Among solids this mode of electron localization is unique to helium. In a comparison of the electron mobility in solid helium to other rare-gas solids, the cavity-localized

electron of helium is at least 10^5 times less mobile than the free electrons of argon, krypton, or xenon (Table 1).

In their theoretical treatment of the localized electron in solid helium, Cohen and Jortner⁷ proposed an experiment to increase the helium density in an attempt to raise the localized electron energy level above that of a free electron state. The energetically favored delocalized electrons would be expected to behave in a manner similar to electrons in other rare-gas solids. In particular, their mobility should increase several orders of magnitude. This transition is expected to take place at about 4000 atmospheres pressure according to their calculation. This paper reports on the search for the delocalized electron state in helium.

Table 1. The Electron Mobility in Four Rare-Gas Solids

T_c is the critical temperature, T the temperature at which the measurement was made. Pressure was 1 atm. except for helium which was measured at 38 atm.

Species	T_c ($^{\circ}$ K)	T/T_c	μ ($\text{cm}^2/\text{V-sec}$)	Reference
Xe	289.8	.54	4.5×10^3	9
Kr	209.4	.54	3.7×10^3	9
A	150.7	.54	1.0×10^3	9
He ⁴	5.20	.47	$\geq 10^{-5}$	6

EXPERIMENTAL METHOD

The experiment consists of a series of electron mobility measurements at low temperatures and high pressures. A null current method, after Cunsolo,¹⁰ is used to monitor any pressure-induced mobility change. A glass or other nonconducting sample vessel has an extremely limited capacity for pressure containment and is not suitable for the present purpose. A fully hardened beryllium-copper vessel is used. The simple linear array of electrodes commonly used with the null current measurement of carrier mobility is not practical with a metal vessel. A cylindrical electrode configuration in which the pressure vessel becomes a guard electrode is successfully employed.

The gas pressure generating apparatus of the pressure system, similar to that of Goldsmith and Heard,¹¹ is separated by a high pressure valve from the sample vessel and pressure gauge (Figure 1). A 100,000 psi Heise gauge with rated accuracy of ± 100 psi is used to measure pressure. The sample vessel, constructed of hardened beryllium-copper, (Berylco 25 alloy, The Beryllium Corp., Reading, Pa.) is sealed at its closure plug with a brass Bridgman extrusion ring. The mobility probe is mounted directly on the closure plug. The high pressure solid helium sample is frozen

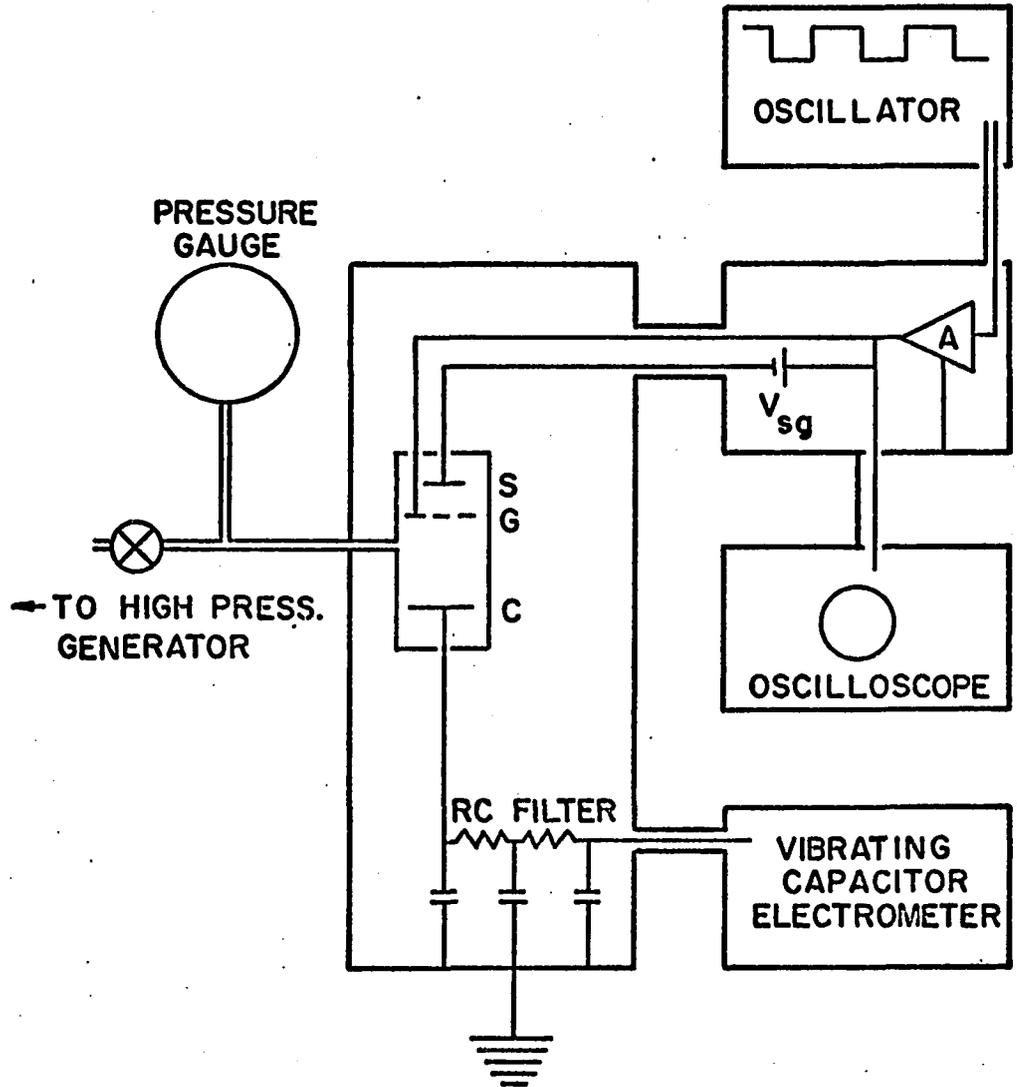


Figure 1. Schematic Diagram of the Apparatus.

isochorically from pressurized helium gas generated at room temperature. At the highest working pressures less than 10% of the generated pressure is lost in lowering the vessel temperature from 300°K to 4.2°K.

A thin cone of EC 2850 GT epoxy (Emerson and Cuming, Inc., Canton, Mass.) is used in place of the standard pipe-stone or lavite cone for electrical lead insulation. This substitution allows all the insulated electrical leads to be sealed in a single cone. No detectable leak is present when the system is isolated by the valve and the vessel filled with solid helium. A vessel filled with gaseous helium leaks slowly through the epoxy at high pressure. This leak, however, is of little experimental consequence.

The current probe (Figure 2) consists of three concentric cylindrical electrodes, the source, grid, and collector. These are mounted concentric with the pressure vessel cavity. The vessel serves as a guard electrode. The electrodes are separated by supports made of Hysol CP2-4289 epoxy (Hysol Corp., Olean, N. Y.). Both epoxy insulating materials are chosen for their high volume resistivity. (Hysol CP2-4289: 6×10^{14} ohm-cm at 298°K; EC 2850 GT: 5×10^{16} ohm-cm at 298°K.) The collector electrode is gold plated copper. The grid and source electrodes are made of brass and copper, respectively. Connections to the

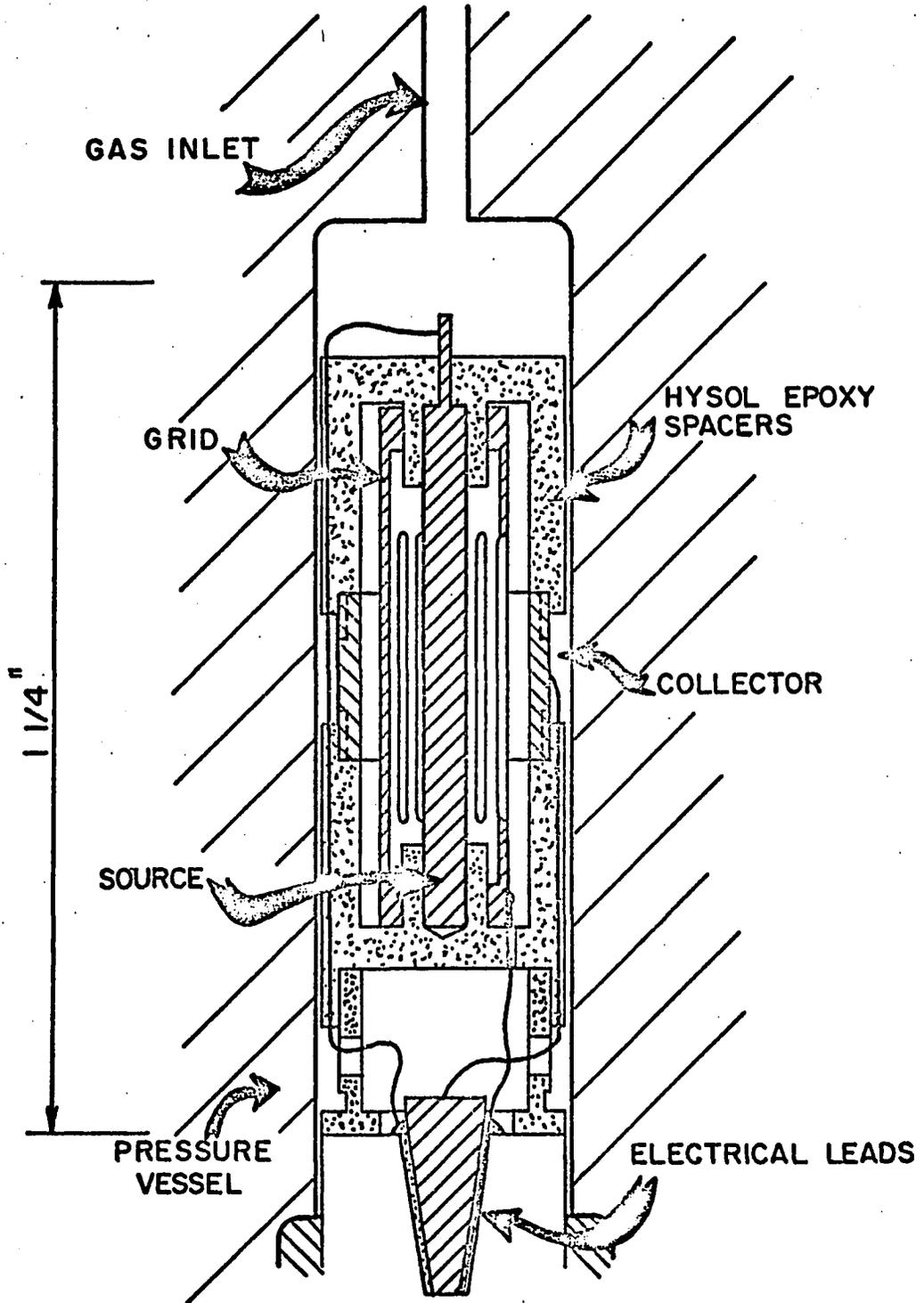


Figure 2. Cross Sectional View of Assembled Probe.

electrodes and the entire electrical network are made with low thermal solder.

The flux of excess electrons is provided by ionization of helium atoms in the vicinity of the source electrode by alpha particle radiation from 10 μc of polonium 210 plated on the source electrode. In solid and liquid helium the range of the 5.30 Mev alpha particles is less than 0.3 mm.¹ This insures that free electrons are not created in the drift space between the grid and collector. Both positive and negative ions can be extracted to the grid depending on the field polarity applied to them.

Temperature is maintained by immersing the high pressure vessel in a pumped bath of liquid helium which is kept stable to within 0.05°K during measurement. Temperature regulation is provided simply by controlling the bath vapor pressure with the pumping speed. A calibrated germanium resistance thermometer is used to measure bath temperatures which can be varied between 1.2 and 4.2°K .

SAMPLE PURITY

The problem of sample purification in this type of measurement has received extensive consideration by many investigators.^{9,12} The operation of the gas high pressure generating apparatus inevitably introduces impurities and one cannot obtain helium purity as high as that attainable in low pressure measurements. Precautions are taken to minimize oxygen contamination of the system, primarily by keeping an overpressure of high purity argon or helium in it at all times. The solid helium is frozen from 99.9999% pure helium gas as purchased from Matheson Gas Products. (East Rutherford, N. J.). To purge contaminants from the pressure system it is alternately filled to tank pressure and bled back to 50 - 100 psi six times before it is pumped to working pressure. In the high pressure runs the gas is pressurized at room temperature and the pressure vessel and gauge are isolated from the rest of the system by the line valve before cooling. Before the sample is cooled to solidification an electric field of reverse polarity is applied between the source and grid electrodes. This procedure removes charged impurities such as oxygen molecules which capture electrons, and it generally results in significantly lower background current levels.

PROBE CALIBRATION

Accurate determination of the electron mobility requires evaluation of the effective probe dimensions as well as determination of the carrier transit time, T_o , between the grid and collector in a known field, E_g . To maximize the probe sensitivity the grid-collector separation is minimized. Thus, of the two radii (i.e., grid and collector) which characterize the drift space, only the collector radius, r_c , can be determined directly. The grid is a thin walled cylinder with slots along its length; the effective grid radius lies within the thickness of its walls. Two grids have been built and used successfully. Grid I has a wall thickness of 0.05 cm which is twice the separation between the outside diameter of the grid and the collector. Comparison of the measurements taken in liquid helium with published mobility data yields an effective grid radius of

$$r_g = 0.257 \pm 0.002 \text{ cm} .$$

This gives a drift distance for this grid as

$$d_I = 0.061 \pm 0.003 \text{ cm} .$$

Grid II is similar to grid I except that its wall thickness is 0.010 cm. A still thinner wall does not offer sufficient

structural rigidity. Calibration results for this grid are shown in Figure 3. The effective grid radius is determined to be

$$r_g = 0.303 \pm 0.001 \text{ cm} .$$

This gives a drift distance

$$d_{II} = 0.015 \pm 0.002 \text{ cm} .$$

In performing the probe calibration experiments in liquid helium satisfactory pressure can be generated by the helium supply tank. In these experiments the pressure generating equipment is bypassed and the helium tank is connected to the high pressure gas line entering the vessel. For work in the liquid the apparatus is limited to detection of transit time T_o below 0.25 seconds by the output RC filter network and the power supplies. The response time with this equipment configuration is less than 1 minute. Alterations to the RC filter network to increase the transit time detection limit to a maximum of 2.5 seconds have resulted in a response time of nearly 30 minutes. This latter arrangement has been tested in liquid helium to confirm its accuracy and is used in solid helium only with grid II.

With a cylindrical electrode probe the mobility of an ion is given by

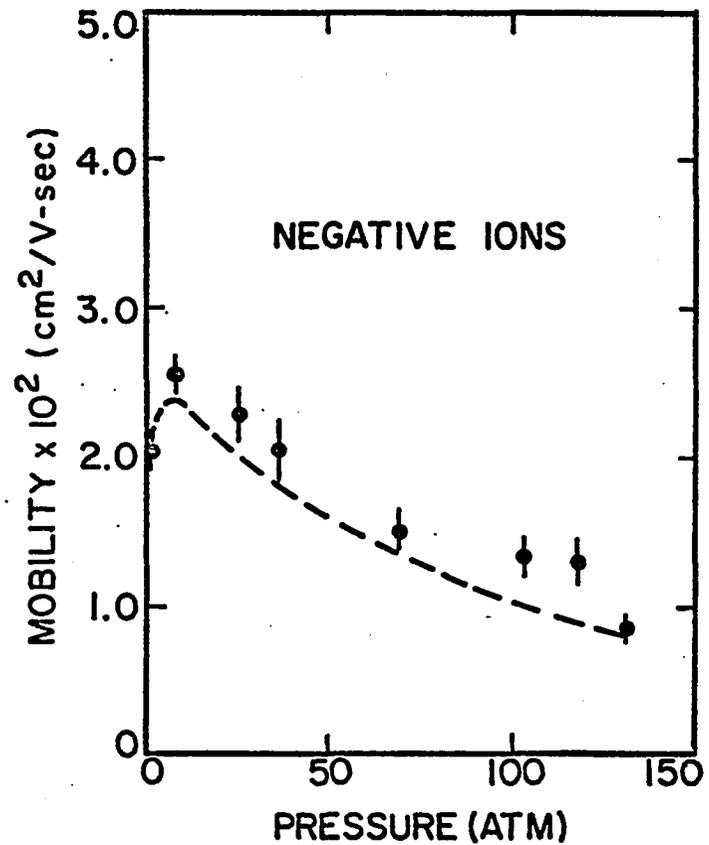
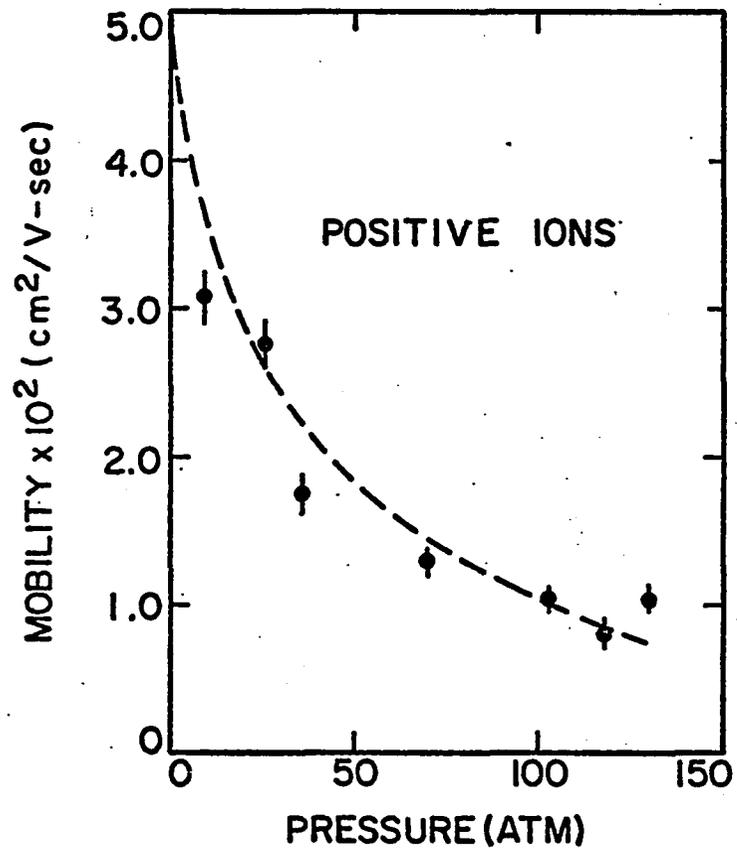


Figure 3. Calibration Results for Grid II.

Ion mobility versus pressure, 4.2°K. Dashed curves from Keshishev, Mezhov-Deglin, and Shal'nikov.⁶

$$\mu = \frac{1}{2T_0 V_g} (r_c^2 - r_g^2) \ln \left(\frac{r_c}{r_g} \right)$$

(See Appendix A.) This expression can be used to establish the sensitivity of the equipment. That information is listed in Table 2.

Table 2. Apparatus Sensitivity

The apparatus sensitivity is listed as the lower detectable limit of the mobility for three equipment configurations.

Grid Design	Response Time (minutes)	Apparatus Sensitivity (cm ² /V-sec.)
I	1	3.0 × 10 ⁻⁴
II	1	1.6 × 10 ⁻⁵
II	30	1.6 × 10 ⁻⁶

EXPERIMENTAL RESULTS

Electron mobility measurements in high density helium at low temperatures have been performed over a wide pressure range. The measurements in solid helium are supplemented by isolated measurements in the liquid at very low temperatures and in the dense gas at a temperature of approximately 75°K.

In solid helium the probe sensitivity is 1.6×10^{-6} cm²/V-sec. At 4.2°K no charge transport has been observed at pressures up to 6400 atm in a maximum field of 3300 V/cm. Similar measurements at 1.2°K and 32 atm yielded the same result. Two experimental runs have been performed at high pressure in which the temperature of the helium sample was allowed to rise above the melting temperature from 4.2°K. In both cases fast probe response is needed to follow the warm-up, thus necessitating use of probe sensitivities an order of magnitude poorer than that attainable with the slow probe. The first run with a sensitivity of 3.0×10^{-4} cm²/V-sec terminated with a pressure of 6270 atm at the melting point of 45°K, and the second with a sensitivity of 1.6×10^{-5} cm²/V-sec melted at 48°K with a pressure of 6660 atm. Electronic charge transport was not observed in these runs at any temperature below the melting point. The

present result, in spite of comparable probe sensitivity, does not agree with the results obtained by Keshishev et al.⁶ who report the negative ion mobility value of 9.7×10^{-6} cm²/V-sec in a field of 1.52×10^4 V/cm at 1.96°K and 38 atm.

Measurements in liquid helium at temperatures below 4.2°K are in satisfactory agreement with work published by other experimenters. These results are reproducible when the sample phase is cycled from liquid to solid and back to liquid, indicating that no damage has been sustained by the probe during the phase changes. Results are shown in Figure 3; a typical raw data plot of collector current versus frequency is shown in Figure 4.

The results of mobility measurements made in the dense gas at 6760 atm and $75 \pm 5^\circ\text{K}$ of both the positive and negative charge carriers are given below.

$$\mu_- = (3.1 \pm 0.5) \times 10^{-3} \text{ cm}^2/\text{V-sec}$$

$$\mu_+ = (4.9 \pm 0.6) \times 10^{-3} \text{ cm}^2/\text{V-sec} .$$

The large uncertainty in the temperature results from a lack of sensitivity in the germanium resistance thermometer in this temperature range and from lack of provision for thermal stabilization above liquid helium bath temperatures.

The experimental data for these results are shown in

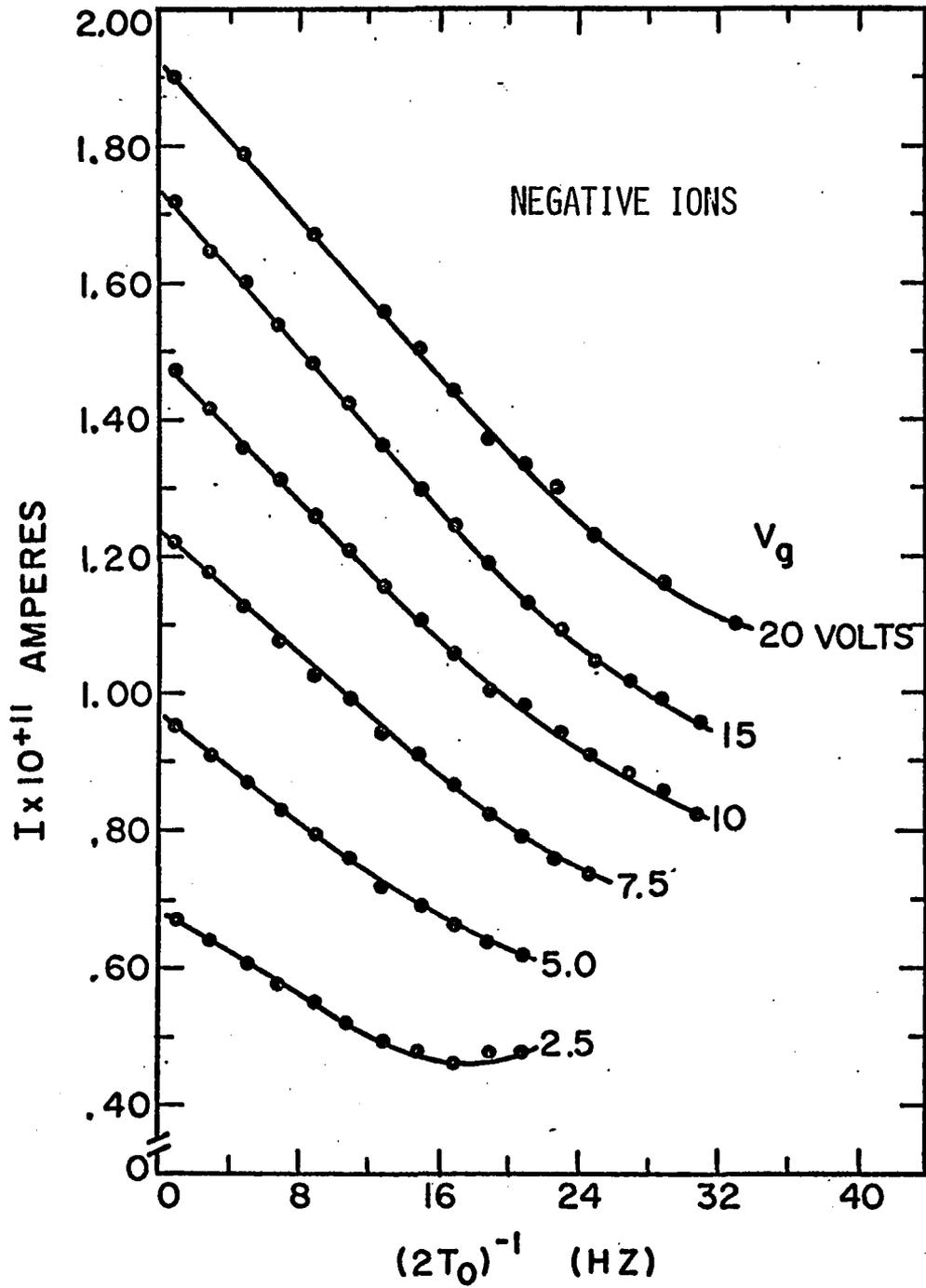


Figure 4. Typical Raw Data for Liquid Helium.

Figure 5. The relatively large background currents are attributed to contamination of the probe by the polonium-210 isotope. The voltage dependent structure of the curves is attributed to stray electrostatic fields. Although such structure is not evident in the data taken in liquid helium, it has been seen when the grid-source potential is low.

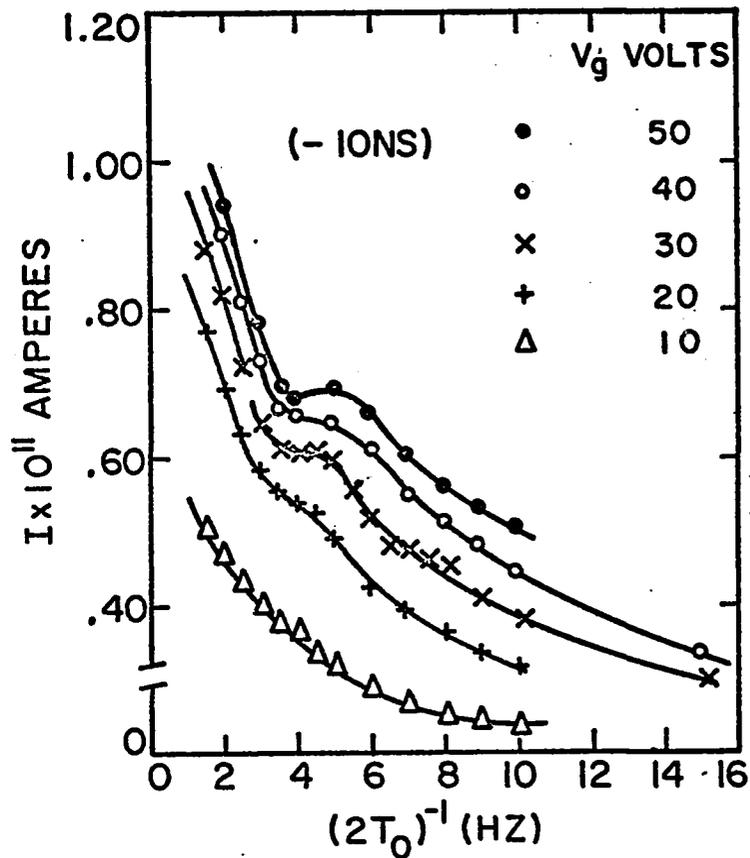
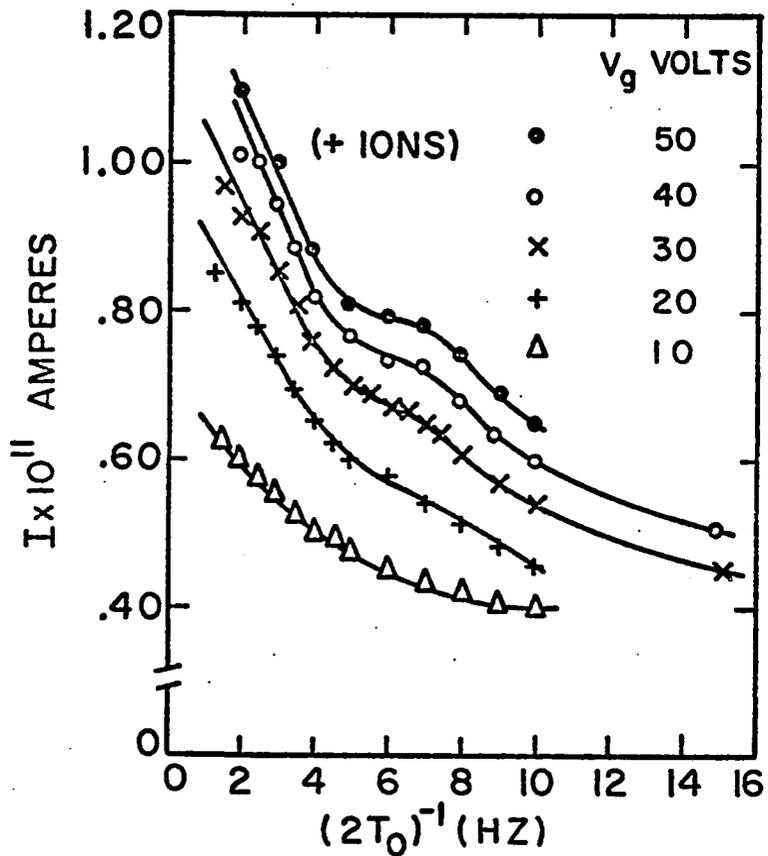


Figure 5. Raw Data for Dense Helium Gas.

Pressure 6760 atm.

Temperature $75 \pm 5^\circ\text{K}$

DISCUSSION

Two explanations may account for the results of the present experiments and deserve consideration. First, the localized electron state may not collapse at or below the applied pressure. Second, the electrons may be trapped whether they are cavity localized or quasifree. Either description is consistent with the data and each will be discussed.

Let us first assume that no ion trapping mechanisms of sufficient strength or number are present in the solid helium sample. Earlier work has confirmed the presence of cavity-localized electrons in the solid phase at low pressures.^{6,7} The existence of localized electron states at the maximum pressure applied is indicated by both direct and indirect experimental results. Primary evidence is given by the fact that no high-mobility ion was detected in the solid below its melting temperature. Indirect evidence comes from analysis of the present results in the dense gas. It is shown below that electron bubbles with a radius of approximately 6 Å exist in the gas at densities similar to that in 4000 atm solid helium. Since helium does not undergo a large volume change on melting (< 5%) it is most probable that similar sized bubbles exist in the solid near the

melting point at this pressure. Thus the present observations are consistent with the presence of cavity-localized electrons in the solid at the highest experimental pressures, nearly 7000 atm, which are approximately 75% larger than the estimation offered by Cohen and Jortner.⁷

The size of the negative ion in the dense gas can be estimated in the following manner. The classical hydrodynamics expression for the mobility of a charged solid sphere in a viscous fluid is given by¹³

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

where e is the electronic charge, η is the viscosity of the fluid, and R is the radius of the sphere. One can determine the viscosity from this equation by using the measured value of μ for the positive ions at this pressure and temperature and an estimated value of R . The radius of the positive ion is calculated using Atkins' model¹⁴ of positive ions in liquid helium which takes into account the effect of electrostriction. According to his model this radius is given by

$$R_+ = \left(\frac{N\alpha e^2}{2V_M \epsilon_0^2 (p_s - p)} \right)^{\frac{1}{4}},$$

where $N\alpha$ is the molar polarizability, p_s the solidification

pressure, p the external pressure, V_M the molar volume, and ϵ_0 the permittivity of vacuum. The polarizability is $N\alpha/V_M = 6 \times 10^{-3}$ coul²sec²/gm-cm³ at this pressure.¹³ This gives

$$R_+ = 3.9 \times 10^{-8} \text{ cm .}$$

We can thus calculate a viscosity of the dense helium gas at 75°K as

$$\eta = 4.4 \times 10^{-4} \text{ poise .}$$

At the same time, from μ_+ , μ_- , and R_+ one can obtain the bubble radius as

$$R_- = 6.2 \times 10^{-8} \text{ cm .}$$

Alternatively, one can use the Nernst-Einstein equation to calculate the self-diffusion coefficient D as

$$D = \frac{kT\mu_+}{e} ,$$

where k is Boltzmann's constant and T is the absolute temperature. This in turn can be used with an expression from the significant structure theory of liquids¹⁵ which relates viscosity to the diffusion constant to obtain

$$\eta = \frac{kT}{6[(\sqrt{2}V_M)/N]^{1/3} D} = 2.0 \times 10^{-3} \text{ poise .}$$

This value is expected to be high since we have neglected the effects of electrostriction. The value of the viscosity in normal liquid helium, $\eta = 2.0 \times 10^{-5}$ poise, is expected to be lower than that in the dense cold gas. These values bracket the expected viscosity value. The first number we have calculated above falls close to the center of this range.

The density of helium gas has been estimated in two ways. First, using the molar volume measurements of Bridgman¹⁶ in helium gas at 65°C and the PVT data of Dugdale,¹⁷ one can scale the reduced molar volume as a function of the reduced temperature to obtain the value $V_M = 9.1 \text{ cm}^3/\text{mole}$. Second, one can calculate V_M using a van der Waals-like equation of state in which the molecular cohesion term is neglected,

$$p = \frac{NkT}{V_M - b} .$$

For b we have used the molar volume of the solid at 7000 atm. This, too, gives $V_M = 9.1 \text{ cm}^3/\text{mole}$. The density for this molar volume is $\rho = 0.44 \text{ gm/cm}^3$ which is similar to that in solid helium at 4000 atm.

To summarize, we have assumed that the effect of ion trapping is negligible in the solid helium sample and shown as a result that cavity-localized electrons persist to

pressures near 7000 atm and at all temperatures below the melting point of the solid. This pressure is well in excess on the minimum collapse pressure calculated by Cohen and Jortner of 4000 atm.

Let us now reexamine their solution to the problem of excess electrons in solid helium to see if a more accurate critical pressure can be calculated using the present results. Consider the energy of the cavity-localized electron in the solid. Following Cohen and Jortner,⁷ it can be divided into three terms.

1. A ground state electronic energy term is obtained from the solution of the eigenvalue problem of an electron in a spherical cavity in a nonpolar continuum fluid as¹⁸

$$E_e = X^2 V_0 ,$$

where X is the solution of the boundary condition accounting for the abrupt density discontinuity at the cavity edge and is determined by

$$\cot (Xk_0 R_0) = - \frac{(1 - X^2)^{\frac{1}{2}}}{X} .$$

V_0 is the lowest energy state of the non-localized electron in the solid. Here R_0 is the bubble radius and k_0 is the ground state wave number. V_0 is evaluated by using the Wigner-Seitz method with a helium atom model suggested by

Jortner et al.¹⁹ in which the atom is replaced by a hard sphere of radius a equal to the electron scattering length. The solution yields an electron wave function

$$\psi = \frac{\sin k_0(r - a)}{r} .$$

Application of the boundary condition at the surface of the Wigner-Seitz cell provides a relation between k_0 and the cell radius r_s :

$$\left(\frac{\partial \psi}{\partial r} \right)_{r_s} = 0$$

which yields

$$\tan k_0(r_s - a) = k_0 r_s .$$

The lowest energy is given by

$$V_0 = \frac{\hbar^2 k_0^2}{2m} .$$

2. A pressure-volume term representing the work done against the external pressure p in creating the bubble is given by

$$E_{pV} = \frac{4}{3} \pi R_0^3 p .$$

3. A surface term relating the specific surface free energy F_s to the bubble creation work is given by

$$E_s = 4\pi R_o^2 F_s .$$

Experimental values for F_s at high pressure in solid helium are not known; however, in writing the expression for the pressure-induced collapse criterion below we will assume that E_s is negligible compared to $E_e + E_{pV}$. This follows from the use of the rough estimate of F_s by Cohen and Jortner⁷ which gives $F_s = 8 \times 10^{12}$ ergs/cm² in the present case. Now the total energy of the cavity localized electron is given by

$$E_t = X^2 V_o + \frac{4}{3} \pi R_o^3 p + 4\pi R_o^2 F_s .$$

The criterion that the bubble be stable is

$$E_t < V_o .$$

Let us assume the solid helium structure is close-packed. The Wigner-Seitz method allows calculation of V_o as a function of pressure (Figure 6). One uses the low energy electron-helium scattering length $\underline{a} = 1.13$ a.u. (= 0.60 Å) for the core potential radius. From this value of V_o the electronic and pressure-volume energy terms can be calculated as a function of R_o for a given pressure. These are shown in Figure 7 for a pressure of 6400 atm. The potential has a minimum of 2.5 ev at a bubble radius of 4 Å. If the surface energy is small, the bubble will be stable at this

Figure 6. Pressure Dependence of the Wigner-Seitz Calculation of V_0 .

Shown is the pressure dependence of the Wigner-Seitz calculation of V_0 for 3 different core radii: 0.6 Å, 0.5 Å, and 0.4 Å.

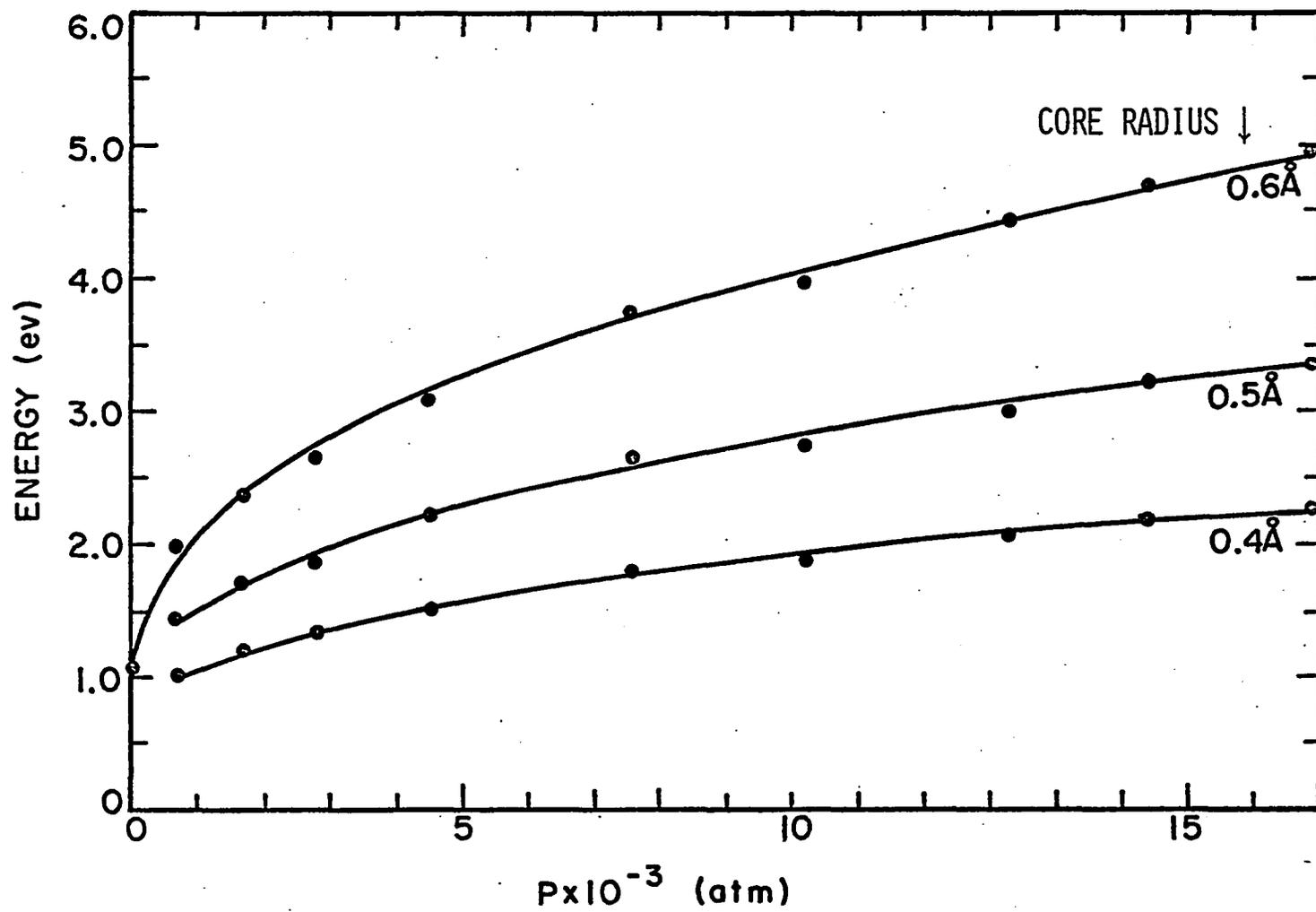


Figure 6. Pressure Dependence of the Wigner-Seitz Calculation of V_0 .

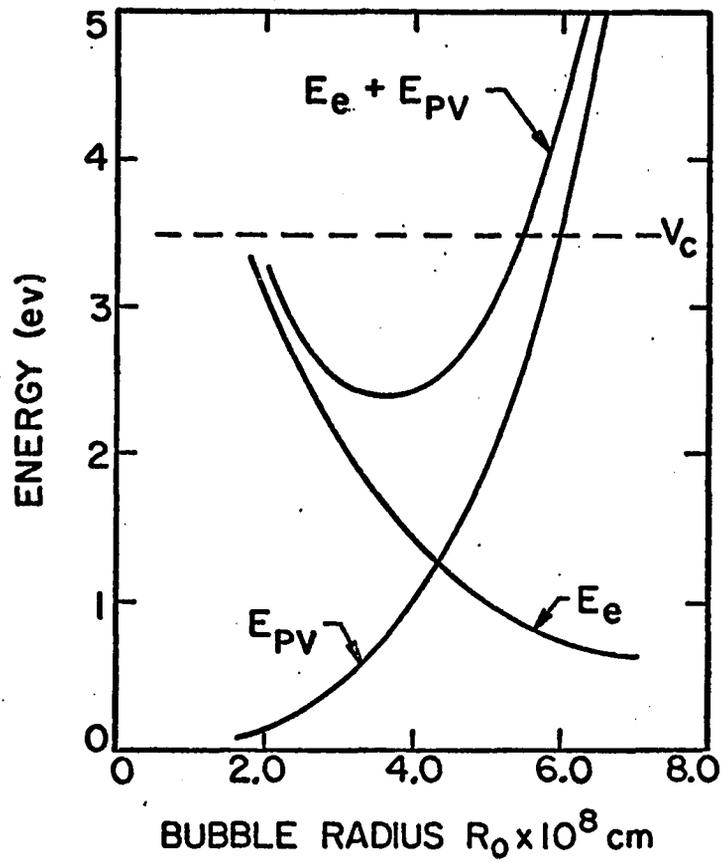


Figure 7. $E_e + E_{pV}$ versus R_0 at 6400 atm.

pressure. When one includes the surface term, bubble stability is assured for $F_s \leq 80 \times 10^{12}$ ergs/cm², an order of magnitude greater than its estimate by the above authors (Figure 8).

The pressure-induced collapse criterion for the cavity-localized electron state can be written as

$$p \geq \frac{3}{4\pi} (1 - X^2) \frac{V_0}{R_0^3} .$$

When one adopts a reasonable bubble radius in the range of 4 - 7 Å (the bubble radius in liquid helium at pressure near solidification is 9 Å) and a wave vector

$$1 \times 10^8 \leq k_0 \leq 1.4 \times 10^8 \text{ cm}^{-1} ,$$

this expression reduces to

$$p \geq 720 V_0^{5/2} ,$$

where p is expressed in atmospheres and V_0 in electron volts. Using the calculation of V_0 made above, we find that the bubble does not collapse at any critical pressure. It is clear from Figure 6 that V_0 is only weakly dependent on p at high pressures while the pressure-volume term in E_t is directly proportional to p . At sufficiently high pressures E_t must exceed V_0 and the non-localized electron state would be favored. The evident weakness of the theory is the

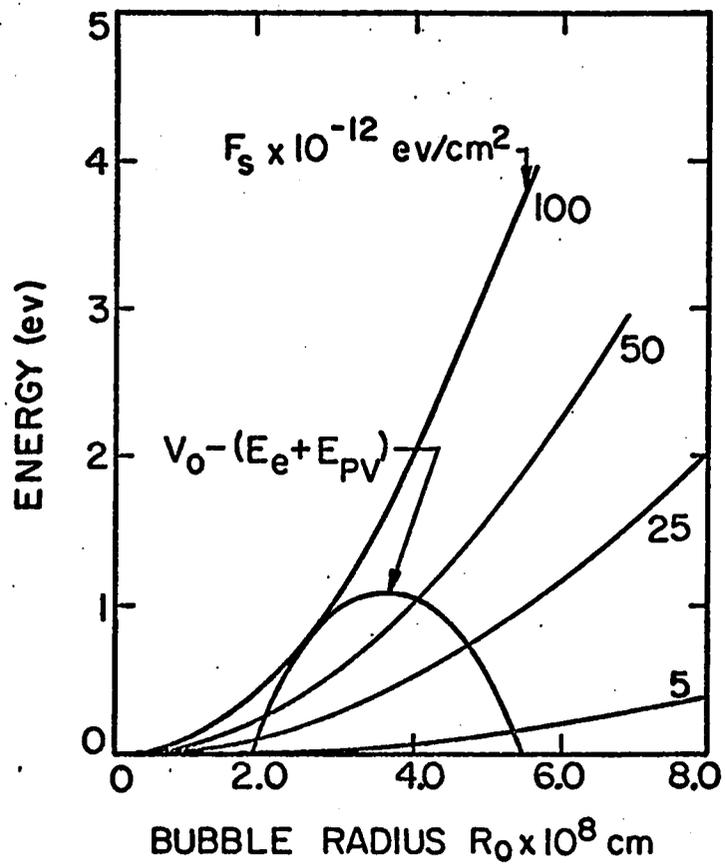


Figure 8. Surface Energy for Several Values of F_s .

estimate of V_0 . The Wigner-Seitz calculation employed here gives an upper limit for this energy because of the restriction on the core size. In the present case with a small bubble radius the localized electron wave function will have non-negligible higher energy components which express themselves by penetrating the atomic potential more deeply than a similar electron in a larger bubble. One can account for this by changing the core dimension a in the helium lattice model used above. The effect of a smaller core radius on the value of V_0 is shown in Figure 6 as a function of pressure. Although the present computation shows that bubble collapse does not occur at any pressure with a 0.6 Å core, it occurs at a pressure ≥ 9000 atm with a 0.5 Å core and at a pressure ≥ 700 atm with a 0.4 Å core. Let us estimate the cavity localized electron energy values for the highest non-negligible components of the wavefunction by

$$E = \frac{\hbar^2 k^2}{2m} ,$$

where

$$k \approx \frac{\pi}{r_s} .$$

This gives

$$E = 10 - 20 \text{ ev} .$$

The core dimension as represented by the electron-helium

atom scattering length can be estimated from the existing measurements of the electron-helium atom cross-section.²⁰

One obtains

$$0.46 \leq a(E) \leq 0.54 \text{ \AA} .$$

This range of the core radius will assure bubble collapse.

The theory predicts collapse pressures of

$$p \geq 20 \times 10^3 \text{ atm} \quad (a = 0.54 \text{ \AA})$$

$$p \geq 3 \times 10^3 \text{ atm} \quad (a = 0.46 \text{ \AA}) .$$

The estimate of $p \geq 4000 \text{ atm}$ given by Cohen and Jortner falls just above the lower limit here. The strong dependence of the critical pressure on the core size makes it very difficult to establish reasonable estimates.

In the light of the large uncertainty associated with the estimation of collapse pressure, the results presented here are understandable. Although we have applied pressure nearly 75% in excess of the minimum predicted for collapse of the cavity-localized electron state by Cohen and Jortner,⁷ and have failed to see the collapse, it is certainly possible that a 500% excess or more may be needed.

The possibility that the cavity-localized electron states in solid helium have not collapsed at the pressures applied is consistent with the observation that there is no

high-mobility charge carrier in the solid at these pressures. However, it fails to account for the fact that within the sensitivity of the instrument the charge carriers are not detected at all in the solid. The possible causes for this apparent total immobility are insufficient instrument sensitivity, actual immobility of the localized electrons, and trapping of the electron. The last possibility is consistent with either a localized or delocalized electron, and both must be considered, although we have shown the probable existence of the cavity-localized electron at all experimental pressures of the present work. Neither insufficient sensitivity nor actual bubble immobility alone are complete explanations of these data. The work of Keshishev et al.⁶ at higher electric fields and at low pressure indicates that both current and mobility sensitivity might be satisfactory in the present work, and that, at least in very high electric fields, the localized electrons are mobile enough to have measurable speed under optimum conditions. It is probable that some type of trapping is present for localized electrons and possibly even for quasifree electrons.

Structural faults such as vacancies and crystal grain boundaries can exist in the solid as well as charged and uncharged impurities. Furthermore, trapping at the interface between the electrodes and the solid helium has been suggested²¹ as a possible mechanism that would be

highly effective for the cavity-localized electrons. A high density of voids in the solid is a possible source of traps in the vicinity of the source electrode where they could be created by radiation damage from the polonium 210. Trapping on the crystal grain boundaries in the solid is also a possible mechanism. The effectiveness of both the void and boundary traps should be strongly temperature dependent, and both should be effective for localized and free electrons. Considering the wide range of temperature employed for the present work, one should have been able to observe some charge release from the traps at temperatures approaching the melting point. This would be especially so for free electrons. Impurities as traps may be discounted since they should be nearly as effective in the liquid as the solid. Finally, we must consider the trapping mechanism operating at the grid-solid helium interface.

The electrons are created between the source and grid of the probe and pass through the grid with a forward electric field into the drift space. Suppose there is an appreciably large density variation in the solid helium wherever it is in contact with a surface. Since it is known that the penetration energy of electrons into liquid helium is 1.0 to 1.3 eV,^{2,3} it is likely that a potential barrier of a similar magnitude makes penetration of electrons into bulk helium solid energetically unfavorable. If, in passing

through the grid, electrons were caught in such a trap they would enter the grid-source circuit and not be detected at the collector. At the present time we know of no evidence for the existence (nor of the absence) of the necessary density variation attributed to this trap. A trap of this type is not expected to be totally effective with free, highly mobile electrons which would have a large probability of entering the drift space through portions of the grid relatively unaffected by the density variation.

In summary, the proposed trapping mechanisms are expected to operate most effectively on cavity-localized electrons and with only partial effectiveness on quasifree electrons. This is consistent with the first part of the discussion which concludes that the electrons most probably remain cavity-localized at the highest experimental pressures.

Strictly speaking, mobility is only defined in the limit as the electric field strength approaches zero. Although Keshishev et al.⁶ have "measured" the electron mobility at the electric field of 15200 V/cm, they have established only a lower value for the zero-field mobility. It is desirable to make measurements at lower fields to determine the actual mobility. Considering the problems in the present experiment which are associated with the presence of the grid and the use of polonium 210, one can suggest an alternate method to measure the cavity-localized

electron mobility. One could use a two electrode probe with a tritium impregnated titanium beta-source. This would ensure creation of the ions in the bulk of the drift space and minimize the probability of radiation damage to the solid helium. The disadvantage of such a probe is the necessity to account for the effective region in which the cavity-localized electron is created.²² Collector current measurements as a function of electrode spacing might resolve this problem.

APPENDIX A

OPERATION OF THE PROBE

Ions of a chosen sign are directed to the grid by an electric field E_{gs} applied between source and grid. A square wave signal of desired frequency f and amplitude $2V_g$ is applied between the grid and the collector. The collector is held at ground potential by low leakage capacitors in the RC filter of the output. The square wave signal is biased center-zero. Thus the ions in the drift space between the grid and the collector experience a field which alternately directs them toward the collector or away from it as the square wave signal changes polarity. The collected charge is measured by a Keithley vibrating capacitor electrometer.

Consider ions entering the drift space from the grid. Ideally, with a relatively high square wave frequency the ions move only a fraction of the drift distance before the reverse field forces them back to the grid. No current will then be detected. As the frequency is decreased the ions are able to drift further across the space, until some frequency f_0 is reached at which the ions begin to arrive at the collector. As the frequency is lowered past f_0 an

average collector current with a linear frequency dependence is measured. The cutoff frequency f_0 can be determined graphically by extrapolation of the linear region, and is related to the drift time T_0 by $f_0 = (2T_0)^{-1}$.

Let r_g = grid radius

r_c = collector radius

E = electric field strength in the drift space

and μ = ion mobility.

Then for $r_g < r < r_c$, the drift velocity v is given by

$$\frac{dr}{dt} = v = \mu E .$$

For the radial electrode arrangement

$$E = \frac{V_g}{r \ln(r_c/r_g)} .$$

Thus

$$\frac{dr}{dt} = \frac{\mu V_g}{r \ln(r_c/r_g)} ,$$

or

$$r dr = \frac{\mu V_g}{\ln(r_c/r_g)} dt .$$

One integrates this over the time T_0 that it takes the ion to move from the grid to the collector to obtain

$$\frac{1}{2}(r_c^2 - r_g^2) = \frac{\mu V_g T_o}{\ln(r_c/r_g)} .$$

Hence

$$\mu = \frac{1}{2T_o V_g} (r_c^2 - r_g^2) \ln \left(\frac{r_c}{r_g} \right) .$$

This expression gives the ion mobility over the range of voltages V_g in which the ion drift velocity is the linearly increasing function of V_g . At high electric fields, in which the drift velocity is not linearly dependent on V_g , this expression established a lower limit for the actual ion mobility.

REFERENCES

1. F. Reif and L. Meyer, Phys. Rev. 119, 1164 (1960).
2. W. T. Sommer, Phys. Rev. Let. 12, 271 (1964).
3. M. A. Woolf and G. W. Rayfield, Phys. Rev. Let. 15, 235 (1965).
4. H. R. Harrison, University of Michigan Thesis, 1971.
5. J. L. Levine and T. M. Saunders, Phys. Rev. 154, 138 (1967).
6. K. O. Keshishev, L. P. Mezhev-Deglin, and A. I. Shal'nikov, Zh. Eksp. Teor. Fiz. Pis. Red. 12, 234 (1970) in Russian. English Translation in Sov. Phys. JETP Let. 12, 160 (1971).
7. M. H. Cohen and J. Jortner, Phys. Rev. 180, 238 (1969).
8. J. Jortner, N. R. Kestner, S. A. Rice, and M. H. Cohen, J. Chem. Phys. 43, 2614 (1965).
9. L. S. Miller, S. Howe, and W. E. Spear, Phys. Rev. 166, 871 (1968).
10. S. Cunsolo, Il Nuovo Cim. 21, 76 (1961).
11. J. R. Goldsmith and H. C. Heard, J. Geol. 69, 45 (1961).
12. H. Schnyders, S. A. Rice, and L. Meyer, Phys. Rev. 150, 127 (1966).
13. R. G. Arkhipov, Usp. Fiz. Nauk. 88, 185 (1966) in Russian. English Translation in Sov. Phys. Uspekhi 9, 174 (1966).
14. K. R. Atkins, Phys. Rev. 116, 1339 (1959).
15. H. Eyring and M. S. Jhon, Significant Liquid Structures, John Wiley and Sons, Inc., 1969.
16. P. W. Bridgman, The Physics of High Pressure, G. Bell and Sons, Ltd., 1958.

17. J. S. Dugdale, *Nuovo Cim. Suppl.* 9, 27 (1958).
18. B. E. Springett, M. H. Cohen, and J. Jortner, *Phys. Rev.* 159, 183 (1967).
19. J. Jortner, N. R. Kestner, S. A. Rice and M. H. Cohen, Modern Quantum Chemistry, Istanbul Lectures, Ed. O. Sinanoglu (Academic Press, Inc., New York, 1966), p. 129.
20. D. E. Golden and H. W. Bandel, *Phys. Rev.* 138, A14 (1965).
21. R. A. Young, private communication. Department of Physics, The University of Arizona, Tucson, Arizona.
22. R. L. Williams, *Can. J. Phys.* 35, 157 (1957).