CONDUCTION PROCESSES IN LIQUIDS

by

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ABSTRACT

In the work described in this thesis attempts have been made to obtain information on three aspects of the behaviour of liquid argon as an ionization counter. Ionization was produced by alpha particles from a source deposited on the negative electrode of a small parallel-plate chamber in which pure argon was liquefied. The current pulses resulting from movement of the liberated electrons in the field applied to the plates were analysed electronically.

Firstly, it was desired to determine the time taken by the electrons originating from distinct ionization events to traverse measured electrode separations and be collected by the positive electrode. These transit times depend upon the electron mobility in liquid argon, defined as the velocity of the electrons per unit field. From this mobility, the mean free path and collision cross-section of the electrons with respect to argon atoms can be calculated. Some estimates of transit times and mobilities, and resulting values of mean free path and cross-section are given. However, it appears that limitations of the electrode spacing and the applied field cause the transit time to be so short as to necessitate the use of wide band amplifiers with, consequently, high noise levels.

Secondly, the causes of variation in size of current pulses with angle of emission of the initiating alpha particles were investigated. The effect of greatest interest was that due to the geometry of the chamber, as from it a determination was made of the range of alpha particles in liquid argon. For 5.3 MeV alpha particles, the weighted centre of ionization was found to be 0.006 cms. from the source, implying a particle range of about 0.009 cms.

The number of electrons contributing to current pulses was also
found to be a function of the angle of emission, on account of a variable degree of recombination with the positive ion column. This number was determined, and even under the most advantageous conditions fell considerably short of the probable total number liberated. This fact imposes a serious limitation on the potentialities of liquid argon as a useful counter.
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CONDUCTION PROCESSES IN LIQUIDS

INTRODUCTION

The theory of conduction pulses began with the work of Röntgen and Joffé (1913), H. Schiller (1926) and G. Jaffé (1932). The first experimental observation of such pulses was found by Van Heerden (1945) who used single crystals of silver chloride. Since that date much work has been done on both crystals and semi-conductors and the whole field has been adequately surveyed by Hofstadter (1949).

The explanation of the conduction properties of these materials is based upon the theory of filled and empty electron energy bands (Williams, 1952). It is postulated that in a normal crystal nearly all the electrons occupy levels in filled bands and hence may be considered bound to distinct ions in the crystal lattice. When an ionizing particle such as an alpha particle enters the crystal, it gives some of these electrons sufficient energy to enter the conduction band in which they are free to migrate through the crystal, subject to collision with the ions. The holes in the crystal lattice resulting from the removal of such bound electrons can behave as positively charged particles due to the migration of electrons from neighbouring ions to them; this is equivalent to movement of the positively charged sites. In most crystal theories such holes are assumed to be relatively immobile, which is found to be the case in AgCl at low temperatures. On the other hand the electrons in the conduction band will, in the presence of an electric field, move through the crystal toward the positive electrode until they reach it or are stopped. They may be stopped by recombination with holes, or by being captured by trapping centres due to inhomogeneities or impurities in the crystal lattice. Both these effects will decrease the resulting conductivity pulses, trapping centres being the more important.
There is another effect occurring in crystals which renders their use as single pulse counters more haphazard. This is the polarization effect caused by the successive trapping of electrons from a large number of pulses and the subsequent building up of potential barriers at positions fixed by the trapping centres. Simultaneously there occurs an analogous process involving the positive ions formed near the negative electrode. The effect of these two charged barriers is to oppose the external field, thus decreasing the size of subsequent pulses, and in some cases quenching them completely.

Finally, in any crystal, if a cleavage plane or crack is present near the negative electrode it forms an effective barrier to electron penetration, resulting in diminished pulses. Such barriers are present in all mosaic crystal structures including lithium fluoride and the other alkali halides, and are regarded by most researchers in the field to be the reason why these crystals do not act as counters although they have large electron mobilities and hence large mean free paths - a necessary condition for the observation of pulses. (Hofstadter, 1950).

These three disadvantages present in crystals, viz., the presence of traps of the form of vacant lattice sites; polarization due to fixed trapping centres; and cracks forming electron barriers, are absent in a liquid counter. However, there are definite limitations on the types of liquids which can be used. These are set by the following conditions:

1. The resistivity of the substance must be high, e.g. $\geq 10^{17}$ ohm-cms.
2. The liquid molecules should have no electron affinity, i.e. the energy of heavy negative ions should not be less than that of neutral atoms.
3. As mentioned above electrons in the liquid should have a large mean free path. However, this quantity is at present unknown for most liquids.
(4) It should be possible to obtain and maintain the liquid in a high state of purity.

(5) Finally, it is advantageous to use materials with a low dielectric constant.

The substances which best fit these conditions and particularly the second are the rare gases in the liquid state. As argon can be easily liquified (boiling point = -185.7 °C at one atmosphere) and is relatively cheap it has been the one most often used and is the one used here. Besides having none of the three disadvantages mentioned above as present in crystals, liquid argon can be obtained quite pure (99.98%) and therefore with few traps due to the presence of impurity atoms having electron affinity. Pulses of good size have been obtained with it.

An argon counter possesses all the advantages of a crystal over gas counters such as the Geiger-Muller type. These are the fast resolving time, and the high efficiency in gamma counting due to the much greater stopping power of liquids and solids over gases. The liquid also lends itself to the using of more compact construction for counting devices; however, this is somewhat offset by the necessity of working at low temperatures.

In the following investigations it was desired to measure the rise times and amplitude distributions of pulses initiated by alpha particles of various field strengths, in order to determine:

(1) the voltage - electron mobility relationship;

(2) the mobility, and hence the mean free path of electrons in argon and the capture cross-section involved in their removal;

(3) the pulse amplitude distribution and the ionisation current and hence the energy per ion pair.
THEORETICAL CONSIDERATIONS

Theory of pulse production

When an alpha particle enters a medium such as liquid argon it loses energy by ionizing the argon atoms within a small cylinder which is centred about its track in the liquid. The rate of loss of energy of such a particle due to ionization reaches a maximum value near the end of its path, which rate is approximately twice the initial one; the rate of ionization then drops abruptly to zero at the limit of the alpha range. However, as this range is assumed to be short with respect to the inter-electrode distance used here, the assumption will be made, at least initially, that the electrons produced by an alpha particle through ionization are all liberated simultaneously in the plane of the negative electrode. It is assumed that no secondary ionization takes place, i.e., there is no ionization of argon atoms by the electrons as they move towards the collecting electrode. These primary electrons will then move with random velocities due to thermal agitation. When an external field is applied there will be superimposed upon this motion a drift velocity through the liquid in the direction of the positive or collecting electrode. Due to the fact that the neutral argon atom has a negative electron affinity there will be no capture of the electrons by such atoms. Hence the only reduction in the number of electrons reaching the collecting electrode will be due to recombination with positive argon ions and capture by impurity atoms possessing electron affinity. The former process will occur only in the region of the source as we have assumed that there is no secondary ionization by the liberated electrons. The latter process can be rendered negligible by using highly purified argon, with the oxygen content particularly, reduced to a minimum.
In moving toward the positive electrode these electrons cause a current, $i$, to flow for a time, $\tau$, through the external circuit. $\tau$, the transit time for the electrons is given by

$$\tau = \frac{l}{v}$$

where $v =$ the drift velocity of the electrons in the direction of the applied field and $l =$ the electrode spacing distance.

This gives rise to a current pulse of the form shown in figure 1(b). This is an approximation as it will be rounded by a variety of effects, the most important of which are:

1. The difference in distance travelled by electrons liberated near the start of the ionizing particle's range and those liberated at the end for an ionizing track nearly perpendicular to the electrodes. This will be discussed further in a following section. There is also the difference in time of liberation by the alpha as the latter proceeds along its track in the liquid. This allows some electrons to complete their path to the collection electrode before others and results in a widening of the whole pulse. The high velocities of alphas of the energy used here will keep this effect small.

2. Straggling in the times of arrival at the positive electrode due to statistical fluctuations in the number of collisions encountered by various electrons. However, in fields of the magnitude used here ($\sim 10^4$ volts/cm.) this effect is considered small. It therefore is assumed that figure 1(b) represents a current pulse which closely approximates to the true picture.

This current will flow through the external circuit which is given by figure 1(a) with $C$ representing all the stray capacitances arising from the chamber, and leads to the pre-amplifier. $R$ is the grid resistance.
FIG. 1 (a) INPUT CIRCUIT

FIG. 1 (b) CURRENT PULSE

FIG. 1 (c) VOLTAGE PULSE
of the input stage of the pre-amplifier. This external circuit shapes the current pulse into a voltage pulse of the form shown in figure 1(c); this pulse is amplified and displayed on an oscilloscope screen on which its rise time may be measured.

By measuring the rise time of the voltage pulse, it is possible to find the time of transit of a group of electrons moving through the counter medium a distance \( l \) at a velocity \( v \) in the direction of an applied field of \( E \) volts per cm. The electron mobility in a particular medium is defined as the drift velocity of electrons per unit field.

\[
K \equiv \frac{\nu}{E}
\]

Hence the mobility is given as:

\[
K = \frac{l}{E\tau} \text{ cm.}^2/\text{volt-sec.}
\]

In this measurement of \( \tau \) and hence of \( K \) it is important that the time constant of the input impedance, viz. \( RC \), be small with respect to the rise time \( \tau \). This will also be a necessary condition for all electronic circuits following this; in particular, both the pre-amplifier and the amplifier must have uniform amplification over a wide band width.

**Kinetic Theory of Mobility**

The kinetic theory of gases, along with Maxwell-Boltzmann statistics has been applied successfully to liquids for the determination of such quantities as heat conduction and viscosity (see G. Jaffé, 1949). It is hoped that such an approach can yield satisfactory results in the problem of electron mobilities and mean free paths for the motion of electrons in a liquid.

Langevin (1950) has shown that for electrons of mass \( m \), and
charge e, moving through a gas of neutral atoms of mass M the mobility is given by

\[ K = 0.815 \frac{e}{m C_m} \sqrt{\frac{M + m}{m}} \]  

(1)

where \( L \) is the mean free path of the electrons in the gas and \( C_M \) is the root mean square velocity of the atoms.

Since

\[ \frac{1}{2} M C_m^2 = \frac{1}{2} m C_m^2 \]

where \( C_m \) is the velocity of the electrons, assuming no field is acting, equation #1 can be changed to

\[ K = 0.815 \frac{e L}{m C_m} \sqrt{\frac{M + m}{m}} \]

which, as \( m \ll M \) can be simplified to

\[ K = 0.815 \frac{e L}{m C_m} \]  

(2)

If an electric field is present it will act only on the electrons and ions present. Hence the velocity \( C_M \) of the atoms remains the same, but the expression for electron velocity has a term added to it due to the electric field. Then \( C_m \) depends on both \( E \) and \( T \), and the equation for it must now be found. Equation #2 will still be valid, provided that \( C_m \) is replaced by this new field dependent velocity for the electrons.

From the definition of mobility the velocity in the direction of the field can be written equal to \( KE \), and so the average distance, \( S \), which the electrons move between collisions with the atoms (in the direction of the field \( E \)) is given by:

\[ S = KE \frac{L}{C_m} \]

where \( C_m \) is the average velocity of the electrons and is \( \approx 0.922 \ C_m \) by kinetic theory. \( C_m \) now includes both the velocity due to thermal agitation
energy and that due to the electric field. Therefore:

\[ S = 0.75 \frac{L^2 e E}{m \cdot \gamma_m^2} \]  

(3)

from equation #2.

We now define an equivalent potential per unit charge, \( u \), such that

\[ u = \frac{m c^2}{a e} \]  = the energy of the electron due to thermal agitation plus the energy due to the field.

Then

\[ S = 0.441 \frac{L^2 E}{u} \]  

(4)

Compton (1923) showed that the fraction \( f \), of the average energy lost in an electron-atom collision is:

\[ f = \frac{8}{3} \frac{M m}{(M+m)^2} (1 - \frac{\Omega}{u}) \]  

(5)

for \( \Omega = \frac{3kT}{2e} \) = the energy of the electron per unit charge due to the thermal agitation. This will hold only for fields large enough to give \( u \gg \Omega \). Therefore the change in energy, \( \delta u \) in a distance \( \delta x \) moved by an electron in the direction of a field \( E \) is given by:

\[ e \delta u = e E \delta x - \frac{e u \delta x}{S} \]

or

\[ \frac{du}{dx} = E - 6.02 \frac{m u (u - \Omega)}{M (L^2 E)} \]  

(6)

from equations #4 and #5, with \( M \gg m \).

By assuming that a Maxwellian distribution gives a good approximation to the electron velocities the above expression in \( du/dx \) can be integrated, giving the following equation for \( u \) as a function of \( x \):
\[
\begin{align*}
    u_x &= \frac{n}{2} + E \left[ \frac{1}{\alpha^2} + \frac{n^2 e^2}{4e^2 \alpha^2} \frac{2e^2 \sqrt{\frac{1}{\alpha^2} + \frac{n^2}{4e^2} \alpha^2} x}{\alpha^2 \sqrt{\frac{1}{\alpha^2} + \frac{n^2}{4e^2} \alpha^2} + 1} \right]^{1/2} \\
    \alpha^2 &= \frac{6.02m}{L^2M}
\end{align*}
\]

Now as \(x \to \infty\) the exponentials in equation \#7 both \(\to \infty\), the -1 and +1 in the numerator and denominator respectively can be neglected, and the exponentials cancel out. This gives a terminal value to \(u_x\) which is independent of \(x\) and depends only on the field, temperature and mean free path of the electrons. The distance, \(d\), after which \(u_x\) has obtained a fraction \(\phi\) of its terminal energy, \(u_T\) say, is given by the expression:

\[
d = \frac{1}{2a^2} \ln \frac{1 + \phi}{1 - \phi}
\]

For \(L \sim 10^{-6}\) cms, \(1/a\) is \(\sim 10^{-5}\) cms, and so \(d\) is very small even for \(\phi = .99\). This means that the terminal velocity of an electron is reached very soon after it is liberated, and for all practical purposes we can take \(u_T\) as the energy of the electron per unit charge for the whole transit time of the electron. Hence by letting \(x \to \infty\), equation \#7 becomes

\[
u_T = \frac{n}{2} + \left[ \frac{n^2}{4} + \frac{L^2ME^2}{6.02m} \right]^{1/2}
\]

This gives \(C_m\) from the definition of \(u\) where \(C_m = \sqrt{\frac{2e u_T}{m}}\), and this value of \(C_m\) may be substituted into equation \#2, giving for the mobility:

\[
k = \frac{0.815 eL}{m^{1/2} \left[ \frac{3}{2} \sqrt{\frac{3}{4} kT + \frac{L^2 E^2 M}{1.5m^2}} \right]^{1/2}}
\]

Solving this for \(L\):

\[
L = \frac{3}{2} K \left( \frac{2m}{e^2} \right)^{1/2} \sqrt{\frac{kT}{\alpha^2} + \frac{E^2 M K^2}{3}}
\]
This equation will hold only for fields such that \( \mathcal{N} \ll \mathcal{U} \). In equation \#8, \( k^T \approx 10^{-14} \) ergs for \( T = 87^\circ A \), and \( M \approx 10^{-21} \) gm. for argon atoms. Hence for fields of \( 10^3 \) volts/cm. or greater and values of the mobility \( > 10\text{cm}^2 \text{ per volt-sec.} \), the second term under the root sign is nearly 10 times as large as the first. Thus, as the equation is valid only for large fields, for which the contribution of temperature to mean free path is negligible with respect to the contribution from the field, we may contract \#8 to:

\[
L = \left( \frac{1.5 m M}{e^2} \right)^{1/2} K^2 E
\]

This is the same form of equation as that used by Malkin and Schultz (1951) except for the numerical constant. They also assumed that a liquid may be treated as a gas at high pressure and that the mean free path is a constant. Their final equation was:

\[
L = C \left( \frac{m M}{e^2} \right)^{1/2} K^2 E
\]

but they had no way of evaluating \( C \) which they took to be in the range \( 2.3 \leq C \leq 4 \) compared with the value \( \sqrt{1.5} \) or 1.22 used here.

Finally there is the equation for the atomic radius, \( r \), which is given by

\[
r = \sqrt{\frac{1}{\pi LN}}
\]

where \( N \) is the number of atoms per unit volume. The collision cross-section is then taken as simply the cross-sectional area of the atoms with which the electrons collide. It is thus inversely proportional to \( L \) and to \( KE^2 \). Unlike the gaseous collision cross-section, this quantity for a liquid should be constant with respect to the field, just as \( L \) is. This theory indicates that \( K^2E \) must be a constant, or that
in order to give a constant value for the mean free path for all fields greater than a certain minimum field of \( \sim 10^3 \) volts/cm.

This theory has the flaw that all calculations of mean free path and collision cross-section depend solely on a measurement of the mobility from the transit time of the electrons. Thus there can be no alternative and independent method of finding these quantities and so checking the validity of the assumption that a liquid behaves as a gas at high pressure at least as far as electron conduction is concerned. However, one result of this theory is that the mobility is not a constant but varies as \( E^{-\frac{1}{2}} \). This will provide at least one valuable check on whether kinetic theory essentially derived for gases, can be applied to any extent to a liquid. Malkin's results (1951) do indicate this relationship between \( K \) and \( E \), at least in the range of \( 10^4 - 10^5 \) volts/cm.

Discussion of Ramsauer Effect

In the above theory it was assumed, as mentioned previously, that the mean free path was independent of field. This is not the case for a gas, where \( L \) has a minimum value at a particular value of the electric field. This is known as the Ramsauer effect. However, no such minimum has been found to occur for liquid argon. The following is a qualitative argument to account for its absence in dense media such as liquids.

Wave mechanical theory gives for the cross-section of a collision between an electron of wavelength \( \lambda = \frac{h}{\sqrt{2mE}} \) and momentum \( p = \frac{h}{\lambda} = \sqrt{\frac{2E}{m}} \), and an atomic field, the value (Frolich and Mott, 1939):
$$q' = \frac{\lambda^2}{\pi} \sum_{\ell=0}^{\infty} (2\ell+1) \sin^2 \eta_\ell$$

where $\varepsilon$ = the electron energy

$\hbar$ = Planck's constant

$\ell$ = azimuthal quantum number

$\eta_\ell$ = the phase shift introduced in the electron waves by such a collision.

$\eta_\ell$ is dependent upon the scattering potential $V(r)$ and if this is small for a distance comparable to one wavelength we may write for $\eta_\ell$ the following:

$$\eta_\ell = \int \left[ \left( \frac{2\pi}{\lambda} \right)^2 - \frac{2mV(r)}{\hbar^2} - \frac{\ell(\ell+1)}{r^2} \right]^{1/2} dr$$

$$- \int \left[ \left( \frac{2\pi}{\lambda} \right)^2 - \frac{\ell(\ell+1)}{r^2} \right]^{1/2} dr$$

where the lower integration limits are taken to give the integrands a zero limit.

According to Holtsmark (1929) this scattering potential $V(r)$ is large enough in a gas to introduce as many as three additional wavelengths in an electron wave train after collision. This would account for a large variation in collision cross-section observed for gaseous argon. However, in a liquid the inter-atomic distance is so small compared with that of a gas, that an electron at the time of a collision with a particular atom is subject to the potential fields of several surrounding atoms. This potential may be expressed as $U(r)$ which combines the effects due to all the individual atomic fields exerted on the electron at the time of collision. $U(r)$ will introduce a term of value $-2mU(r)/\hbar^2$ into the second integrand in the above expression for $\eta_\ell$. Also, $V(r)$ in the first integrand, will now assume a perturbed value $V'(r)$.

The two integrands above are therefore of the same form, and
differ only in the potential function present in the second term in each. If \( V'(r) \approx U(r) \) the two integrals will cancel giving \( \eta_c \approx 0 \), and so the cross-section becomes independent of electron wavelength and, therefore, of the electron energy. It seems likely that this is the case for liquid argon, and so the collision cross-section will be small and will change little with changes in external field. Thus there should be no Ramsauer type variation of the cross-section for liquids.

**Pulse Size Distribution**

In the theory of pulse production described on page 4, it is assumed that all ionization takes place in the plane of the negative electrode, and that each alpha particle produces \( n_0 \) electrons, all of which contribute to the observed pulses. This would result in a uniform pulse amplitude for all alphas. Such a result is not the case, and four effects cause variation in this amplitude.

The first of these is due to random noise contributions to the pulses. The effect of this noise contribution depends upon the fundamental frequency of the pulses as compared with the band of noise frequencies amplified. If the latter contains a large percentage of high frequency noise, the majority of pulses are recorded at an amplitude greater than their true value, for the noise peaks would cause the pulse to be registered at its maximum height. If the pulses are of a frequency near to the upper half-power frequency of the amplifier, half of them suffer a decrease in amplitude. In general the result of the noise is to increase the amplitude spread of the pulses by an amount dependant upon the signal to noise ratio. The actual spread is difficult to calculate theoretically, but may be corrected for experimentally, as is shown later.

The second cause of amplitude variation is due to the fact that
alphas are emitted at all angles to the plane of the negative electrode. The resultant columns of electrons travel varying distances between the electrodes, depending on the electrode spacing, \( l \), and the alpha range \( R_\alpha \). Davidson and Larsh (1950) discussed this effect, but did not give an equation relating pulse height \( V_\theta \) due to an electron column formed at angle \( \theta \), with the fraction of such alphas emitted at angles between \( \theta \) and \( \theta + d\theta \). We shall attempt to do this as follows:

In fig. 2(a) the number of alpha particles emitted in a region \( d\theta \) about \( \theta \) is given by

\[
dN_\theta = \left( \frac{N_T}{2\pi R_\alpha^2} \right) 2\pi R_\alpha^2 \cos \theta \, d\theta
\]

where \( R_\alpha \) = range of alpha particle

\( N_T \) = total number of alphas emitted per second.

or if \( N_\theta \) is taken to be the number of alphas emitted at angles \( < \theta \), it is given by

\[
N_\theta = \int_0^\theta N_T \cos \theta \, d\theta = N_T \sin \theta
\]  

(1)

If the electrons are considered to be produced uniformly along the track of the alpha, they can be considered centred at \( R_\alpha/2 \). These electrons will move some distance between \( l \) and \( (1 - R_\alpha/2) \) before striking the positive electrode, depending upon the angle of alpha emission. In each case they will give rise to the same current, \( ne/\tau \), in the external circuit, lasting for some average time \( t = \tau - \tau_0 \). Thus the square pulse illustrated in fig. 1(b) becomes that of fig. 2(b). In such a case the total charge transferred by the electrons will vary with \( \theta \), becoming equal to \( ne \) only for \( \theta = 0 \), for which pulse the positive ions now formed adjacent to the negative electrode contribute negligible charge transfer.

Now, for a particular alpha emitted at an angle \( \theta \), the electrons
FIG. 2 (a)  ANGULAR DISTRIBUTION OF ALPHA PARTICLES.

Current

\[ i = \frac{m_e}{e} \]

\[ t_\theta \]

\[ \tau - t_{\nu_2} \]

\[ \tau \]

positive ion currents

Time

FIG. 2 (b)  CURRENT PULSE VARIATION

Voltage

\[ V_{\text{max}} (\theta, \theta) \]

\[ V_{\text{min}} (\theta, \frac{\pi}{2}) \]

\[ \tau - t_{\nu_2} \]

\[ \tau \]

Time

FIG. 2 (c)  VOLTAGE PULSE VARIATION
will traverse a distance, \( 1 - x/2 = 1 - (R/2) \sin \theta \), in a time \( t = T - t_\theta \).

If \( v \) is taken as the terminal velocity of the electrons and is assumed to be reached in a time small with respect to \( t_\theta \), as well as \( T \), these times are given by

\[
T = \frac{L}{v}
\]

and

\[
t_\theta = \frac{R \alpha \sin \theta}{2v} = \frac{R \alpha \tau \sin \theta}{2L}
\]

(2)

The voltage appearing across the resistance \( R \) in fig. 1(c) will rise as the current \( ne/\tau \) flows in the external circuit. If the time constant, \( RC \) of this circuit is large with respect to \( \tau \), the voltage will not reach saturation but will continue to build up until the current drops back to zero. As shown in fig. 2(c), if \( t_\theta \) is small, compared to \( \tau \), the voltage will rise linearly in the region \( \tau - t_\theta \) to \( \tau \), and hence the maximum pulse height obtained will be given by:

\[
V_\theta = a - b \tau
\]

where \( a \) and \( b \) are empirical constants. They may be found from the equation relating the voltage \( V_t \) at any time \( t \) to \( i \), \( R \), \( C \), and \( t \), found from analysis of the input circuit.

The current pulse is given by

\[
i = \begin{cases} 
0 & t < 0 \\
\frac{ne}{\tau} & 0 \leq t \leq \tau \\
0 & t > \tau
\end{cases}
\]

the voltage appearing across \( R \) in fig. 1(c) can be shown to be

\[
V_t = \frac{ne}{\tau} \left( 1 - e^{-t/RC} \right) \quad 0 \leq t \leq \tau \quad (2a)
\]

\[
V_t = \frac{ne}{\tau} \left( e^{\tau/RC} - 1 \right) e^{-t/RC} \quad t > \tau
\]
From the first of these two equation, the maximum voltage $V_\theta$ reached during the pulse is

$$V_\theta = \frac{n e}{c} R \left( 1 - e^{-\frac{\tau - t_\theta}{RC}} \right)$$

Now with $RC \gg \tau$, $e^{-\frac{\tau - t_\theta}{RC}} \ll 1$ and may be approximated to by the first two terms of the exponential series. This gives

$$V_\theta = \frac{n e}{c} \frac{\tau - t_\theta}{c}$$

With equations (1) and (2) substituted into this for $t_\theta$ and $\sin \theta$, $V_\theta$ becomes

$$V_\theta = \frac{n e}{c} - \frac{n e}{c} \frac{R \alpha}{2 \ell} \frac{N_0}{N_T}$$

(3)

Thus the graph of $V_\theta$ against $N_e/N_T$ should be a straight line, from which it should be possible to find both $R$ and $n$, if $c$ is known.

The relationships are

$$\frac{V_{\text{MAX}} - V_{\text{MIN}}}{V_{\text{MAX}}} = \frac{R \alpha}{2 \ell}$$

(4)

$$V_{\text{MAX}} = \frac{n e}{c}$$

(5)

The third cause of pulse size variation is recombination. In figure 3, we have an alpha emitted at an angle $\theta$ to the electrode and surrounded by a cylinder of radius $r$, in which it is assumed that all ionization takes place uniformly across $r$, so that we have an even distribution of $n_0$ positive and $n_0$ negative ions throughout the column. The electrons are then considered to originate along the axis of the cylinder and to travel a distance $x = r \sec \theta$ through it under the direction of the applied field. Each of these electrons sweeps out a volume centred about its track, which will be given by $\sigma^2 x$ where $\sigma^2$ is the capture cross-section of the electron with respect to the positive ions. We then have:
Fig. 3. Alpha Recombination Column
Number of electrons captured = (density of positive ions) x (total volume swept out by \( n_0 \) electrons)

\[
\frac{n_0 \sigma^2}{\pi R^2 R_a} \chi
\]

Therefore the number of electrons escaping recombination and contributing to the current pulse will be given by:

\[
n = n_0 \left(1 - \frac{n_0 \sigma^2}{\pi R^2 R_a} \sec \Theta\right)
\]

Or as \( V_\Theta \propto n \) we should obtain a linear relationship between \( V_\Theta \) and \( \sec \Theta \) of the form \( V_\Theta = a - b \sec \Theta \), applicable to the voltage - number curve obtained after correcting for the first two effects.

Equation, (#6), does not hold for large \( \Theta \). For those electrons originating in the triangle at the end of the alpha track, as shown in fig. 3, the distance travelled in the presence of positive ions will be less than \( x \), due to the finite length of track. This fraction of the total number of electrons increases as \( \Theta \) increases until for \( \Theta = 90^\circ \), all the electrons move through an average distance \( \frac{R}{2} \) in which recombination can take place. However, if \( r \sim \frac{R}{50} \), the distance \( d \), as shown in fig. 3, will be only \( \sim 10\% \) of \( R_a \) for \( \Theta = 80^\circ \), at which point \( N_\Theta \) will be > 98\% of \( N_T \). This will mean an insignificant correction to the above theory for all except the few pulses initiated by \( \alpha \) particles emitted nearly vertically to the plane of the electrodes.

The fourth effect which must be considered, is the variation of \( n_0 \) itself due to variation in alpha energy. This will occur if some alphas originate within the negative electrode instead of at its surface, thus losing some of their energy in the metal instead of in the argon. This may be avoided by proper formation of the source, and it is assumed here that \( n_0 \) is actually a constant for all pulses.
Determination of Ion Pair Energy:

If it is assumed that each alpha particle emitted by the source enters the liquid argon with the same energy, $H$, and forms the same number electron-ion pairs, $n_0$, then

$$n_0 = \frac{H}{\rho} \chi$$

where $\rho$ is the ionization energy of liquid argon. The quantity $\chi$ is the fraction of the energy lost by the alpha in one ionizing event that is actually used to free the electron. The rest of the energy lost appears as the kinetic energy of the electron and ion.

Now $\rho$ has a value of 25.4 ev. for gaseous argon, and this is the maximum value it can have for the liquid. However, if there is in the liquid a conduction or energy band similar to those known to occur in solids, $\rho$ may be considerably less than this, since the ionization energy of the gas is equal to the ionization energy of liquid argon plus the energy necessary to remove an electron completely from the liquid. That conduction bands can exist in liquids is demonstrated by the fact that Ogg (1946) has described such an energy band for liquid ammonia. However, nothing is known of this for liquid argon, and so the ionization potential in the above equation is unknown. Hutchinson (1948) and Gerritsen (1948) as well as Davidson and Larsh (1950) assumed it to be 25.4 ev. However, if $n_0$ and $H$ are known we can find $\rho/\chi$. As $\chi < 1$ this determines the maximum value of the ionization potential; if this is less than 25.4 electron volts, we may accept the idea of conduction band in liquid argon. Actual evaluation of depends upon determining $\chi$. It has a value of 0.6 for NaCl crystals and is thought to be near to this value for other solids; no values are known for liquids.

In order to find $n_0$ it is necessary to find the limit of $n_0$,
the number of electrons collected in the largest pulse for a given field, as the field approaches infinity. Davidson and Larsh (1950) proposed the equation:

$$\ln \left( 1 - \frac{V_{\theta=0}}{V_{\max}} \right) = -\alpha E^{1/2}$$

(1)

with $\alpha$ an arbitrary constant. Then from equation #5 in the previous section it should be possible to calculate $n_0$, corresponding to the $V_{\max}$ in equation #1 above.

Another possible way of evaluating $n_0$ is from the D.C. measurements of electron current as found by an electrometer. This should give

$$I = \overline{n} e N_T$$

where $N_T$ = number of alphas emitted per second;

and $\overline{n}$ = average number of electrons contributing to each current pulse.

This average number of electrons $\overline{n}$, will be different from $n$, due to the variation of recombination with $\theta$. It can be related to $n$ by means of the experimental curves after they have been corrected for noise and variation in pulse height due to $\theta$. Again $\overline{n}$ should vary with $E$ similarly to the variation in $V_{\theta=0}$, and so permit $n_0$ to be found by an independent method.

**DESIGN OF APPARATUS**

**Argon Chambers:**

Three different chambers were used in the attempts to observe argon pulses. All were basically the same, having a pair of plane parallel electrodes with an alpha source deposited on one of them.

The first consisted of a glass dewar flask containing a lucite holder for the brass electrodes, held in place by ceresin wax. The electrical leads entered from below via glass-to-metal seals. The argon was liquefied in a helical glass coil immersed in liquid nitrogen and allowed to flow down
into the dewar; the two were connected by a ball-and-socket connection. This design had several serious drawbacks. The lucite had a large vapour pressure at room temperature which prevented effective degassing of the chamber. The ceresin wax proved very brittle at liquid argon temperatures, and tended to break loose. Most important was the inefficiency of the heat exchange system. Using liquid nitrogen, the argon tended to freeze in the coils and block them, while liquid oxygen necessitated a pressure of 1.3 atmospheres in the system, which was not constructed to withstand it. Furthermore, the liquid argon in the dewar could not be further cooled, and this necessitated keeping the coil constantly immersed in liquid nitrogen throughout the trial.

The second chamber used is shown diagrammatically in fig. 4. It was constructed of copper with the negative electrode and alpha source attached directly to the lid of the chamber. This was fastened to the body of the chamber, by means of screws, tightening down on the "O" ring seal. The collecting electrode was mounted on a kovar seal soldered into the base of the chamber. The chamber itself was surrounded by a brass cylinder attached to the chamber base plate. This was filled with liquid oxygen, and served as an efficient heat exchanger. This in turn was surrounded by styrofoam blocks to provide thermal insulation.

This chamber was used for some time but several drawbacks forced its abandonment. Difficulty in rendering the "O" ring seal vacuum tight at low temperatures caused considerable trouble on a number of occasions, and the electrode spacing could not be easily varied. The most serious objection however was the manner in which the source was held in place under a copper cap with a small hole in it. This placed the source in a recess about 1 mm. deep, and made the electric field about it difficult to determine exactly. It was primarily to eliminate this drawback that a new
FIG. 4. ARGON CHAMBER
The third chamber designed was the one with which all the results were obtained. It was of brass, as it was felt that the slight advantages obtained from copper, pertaining to its thermal and electrical conduction properties, were more than offset by its softness. As shown in fig. 5, it consisted of two electrodes in a small brass cylinder, one of which was situated on a Kovar seal set in the base of the cylinder, while the other electrode was attached to the top by a screw thread. They were adjusted into a plane parallel position with respect to each other, whereupon the copper glass seal was set down and soldered into place. Afterwards the electrode spacing could be easily and accurately varied, the screw pitch on the upper electrode being known. The upper electrode was screwed down until electrical contact was made with the lower electrode and then turned through a measured angle to give the desired spacing. As there were 52 threads to the inch or 2 per mm, it was possible to set a gap of .1 cms. with an error of less than $30^\circ$ or 4%. The liquid oxygen was contained in a glass cylinder attached to the chamber base plate by a groove filled with glycerine. At low temperatures, the glycerine froze to give a tight seal. For the alpha source, a silver wire was snugly fitted through a hole in the lower electrode and smoothed off to be flush with its upper surface. It was upon this that the alpha source was deposited eliminating any odd field effects present in the earlier chambers.

All three chambers were connected to a vacuum system utilizing both a mercury diffusion and a backing pump; these provided a vacuum of better than .001 microns. The pressure was measured by means of Pirani gauge down to .1 microns, and with a McLeod below this; the latter was also used to check periodically the Pirani, which drifted considerably. Initial degassing was necessary, taking from a few hours to a few days to achieve
FIG. 5. ARGON CHAMBER (NATURAL SIZE)
satisfactory vacua in the different chambers. During the degassing the
chamber was heated to about 200°. No degassing was performed between
trials, as argon would be the only gas absorbed.

The argon tank, containing argon of specified purity of 99.98% was
connected to the system through a trap. This latter was cooled with
liquid oxygen prior to liquifying the argon, and was to serve as a trap
for impurities such as water vapour and CO₂ in the gas. It was subsequently
neglected as it proved to be unnecessary. Also added to the original system
was a rubber balloon to provide a safety reservoir of low pressure argon.
However, as it proved impossible to degas and valueless as a safety device,
it was removed.

In all trials with the second two chambers liquid oxygen was used
as the cooling agent. At this temperature (-183°C) the vapour pressure of
liquid argon is 1.3 atmospheres. It was liquified at about 2 atmospheres
which was near the limit of what the stopcocks in the system could take.
The pressure was measured on the gauge attached to the tank and the inflow
of gas controlled by pressure reducing value also attached to the tank.
The latter gave some trouble when degassing until the plastic sealing disc
inside it was replaced by lead.

After trials on the first two chambers, and then several on the
third had failed to yield any true argon pulses, the argon purity was
suspected. According to Davidson and Larsh (1952), the presence of one
part in 10⁵ of oxygen will reduce the pulse height by a factor of 10, while
nitrogen to 1 part in 10³ will reduce the height by 10%. Two methods of
purification were tried. One utilized a solution of pyrogallol and potassium
hydroxide in water. An air trap with stopcocks on top and bottom was placed
in the system between the tank and the chamber and degassed. Just before a trial, the KOH solution was sucked into the bubbler from the bottom, and the pyrogallol in solution added from the top; this was to prevent any atmospheric oxygen from being absorbed by the latter, which is only effective in a strongly basic solution. The argon was then bubbled through the solution, passed through the liquid air trap, and then to the chamber. This should have reduced the oxygen content to well below .001%. However, as no pulses were then detected, and the pyrogallol solution tended to condense in the liquid air trap it was decided to try a second method.

Magnesium will readily combine with oxygen and nitrogen at temperatures of about 500° C; the products are solids with low vapour pressures. Consequently a quartz tube, about one inch in diameter was packed with magnesium turnings held in place by glass wool plugs. The tube was then degassed. During a trial, this tube was heated to about 600° C by a coil wound around it, and the argon was passed slowly over the turnings. At this temperature magnesium, although still a solid, has a vapour pressure of 1 mm., and so the probability of any oxygen or nitrogen molecules colliding and reacting with a magnesium atom should be very high. The gas was passed through the liquid air trap and then to the chamber. At room temperature, magnesium has a vapour pressure \(\sim 10^{-5}\) mm. and so will cause no trouble in the remainder of the system. This purification procedure was continued for some time after pulses were observed; it was then discarded. As no change in pulse size could be observed, it had to be assumed that the argon was already effectively free of these two gases, and attempts at purification were unnecessary.
The alpha particles were emitted from a polonium (radium F) source at an energy of 5.3 MeV; this material has the advantage of yielding a stable end product, viz. lead 206. It was contained in a solution consisting of radium D, E and F in 0.5N HCl. According to Rutherford, Chadwick and Ellis (1912) the polonium can be separated from the other two radioactive elements by two processes.

One method involved the application of a small potential to the solution sufficient to give a current of 25 to 35 microamps/cm. Within these values of current and using two platinum electrodes, only the polonium will be deposited out on the negative electrode. As it will be deposited at a known rate, it should be possible to get sources of the desired strength quite easily. However, with the solution used, it was difficult to keep the current within the prescribed limits; also impurities present in the solution tended to be deposited on the negative electrode as well as polonium. The second method suggested by Rutherford et al was tried simultaneously with the above one. As it was yielding good results it was decided to abandon the first process altogether.

In the second method, use is made of the fact that silver is sufficiently electropositive to displace only the polonium and not the radium D or E from solution. The solution was drawn into a fine glass pipette by means of a piece of rubber tubing closed at one end and attached to the pipette. For use in the first two chambers the source was placed on a thin silver disc about 0.5 cms. in diameter. This disc was coated with glue and allowed to dry; a small hole was then scratched through to the metal. This limited the source to a small area of the silver disc - about 1mm. in diameter. Collodion was also used but was not as effective as the
glue in preventing the source from spreading. The disc was held firmly under the tip of the pipette and the tubing compressed to bring the solution down so that it just touched the metal but did not spread to form a drop. The time for deposition was kept small to prevent migration of the polonium atoms into the silver, and averaged several minutes; this yielded sources of \(40 - 100\) counts per second with negligible beta contamination from the radium D and E. After washing the source thoroughly, it was impossible to reduce source strength by tapping the disc or pressing the source against a second surface. This fact removes the necessity of preventing the electrodes from coming in to contact with each other, as they do when setting the electrode spacing.

For the third chamber the electrode itself was coated with glue, except for the end of the silver wire set through it. Much more care was necessary in this case for both radium D and E will react with the zinc in the brass. It was possible however, to limit contact of the solution with the electrode entirely to the silver. The silver was exposed to the solution for about one minute, yielding a source of about 100 counts per second with negligible beta contamination. By the time results were obtained, this had dropped to \(60 - 70\) counts per second, as the half life of polonium is only 138 days.

Circuit Components

In the observation of the pulses, two considerations govern the choice of the electronic circuits. They are the amplification and time constants necessary in these circuits. Initially the former was underestimated; in the equation giving pulse height in terms of the input parameters and pulse current, a gain of about 100 was considered adequate. Later it was found necessary to change this to at least 400 for visual
observation on an oscilloscope having 1 cm. deflection for .05 input and to better than $10^5$ for scalar readings. The time constants of all components were to be kept $< 10^{-7}$ seconds, as indicated by the mobility values found by Malkin and Schultz in 1951. This necessitated a half power upper frequency limit of at least 10 megacycles.

A Hewlett Packard wideband amplifier (model #460A) was used in all attempts to observe pulses. It consists of two 5 tube distributed amplifier stages arranged in cascade, with an attenuation control inserted on the input to the second stage. Its characteristics include a voltage gain of 3.5 to 10 over a range of frequencies from 500 KC/sec. to about 90 MC/sec., with a rise time limit of $2.6 \times 10^{-9}$ seconds. The noise level is extremely low, around 40 microvolts peak to peak. The input and output impedances are only 200 ohms, and so the Hewlett Packard could not be used connected directly to the chamber as a much higher resistance was needed at the input, to provide a reasonable voltage value for a pulse.

Accordingly, a small 2 stage pre-amplifier with an anode follower preceded the Hewlett Packard amplifier. Employing 6AK5 tubes and shunt peaking coils it provided a further gain of 10 over a pass band extending up to about 10 megacycles per sec. Its input grid resistor was variously set from 500K down to 3K. This amplifier had a high noise level of approximately 1 millivolt; at first this was thought to be unimportant as pulses of 2 - 3 millivolts were expected. Later events and calculations showed this to be in error by a factor of almost 10, and so it was necessary to discard the amplifier in favour of one with a higher amplification and lower noise level.

A five stage amplifier, plus a cathode follower, was designed and constructed to provide a gain of around 400. Shunt peaking was also
employed in it to increase the half power upper frequency limit from 5 megacycles to around 15. This of course resulted in a fairly large noise voltage, about 3 or 4 times the theoretical limit of amplifier noise set by $W_N = kT_B$ giving 66 microvolts zero to peak for an input resistance of 100K. Many attempts were made to reduce this noise, as well as to smooth the amplification curve - it was constant for only a small portion of the pass band, and dropped off slowly below 5 megacycles when shunt peaking was employed.

After all attempts to measure rise times were discarded, and it was decided to concentrate on pulse distribution, there was no need for such a wide pass band. A two stage pre-amplifier, plus two cathode followers, one providing feedback to the 1st stage, the other providing an output signal, had been built to act as a pre-amplifier for an Atomic linear amplifier. The circuit is shown in fig. 6(a). The amplification obtained with it was very constant at a value of 10 up to about 2 megacycles, with its pass band extending from <100KC/sec to 5MC/sec. The noise level was about 90 $\mu$V zero to peak for 100K input impedance, compared to the theoretical limit of 45 $\mu$V.

The final arrangement of circuit components was that shown in fig. 6(b). The linear amplifier, model 204C, has an input time constant of 0.2 $\mu$sec. on the fast range, and employs feedback to further vary the rise time and amplification. It has a high level output, the signal from which was fed into the scaler, and a low level one of 200 ohms output impedance. The scaler was an Atomic, scale of 64, model 101A, which could handle pulses up to 60 volts. The oscilloscope used, a Tektronix model #517, could be connected directly onto the low level, or as was found necessary due to the gain, via a line attenuator of 20 decibels attenuation. For the best setting of time constant and feedback on the
FIG. 6 (a) PRE-AMPLIFIER CIRCUIT

FIG. 6 (b) CIRCUIT DIAGRAM
linear amplifier, viz. that giving the sharpest pulses, the low level gain was 56 times, giving a total gain of $5.6 \times 10^3$ at the low level output, corresponding to 4.5 microvolts per division on the scaler.

The amplifications noted above were all measured with a Triplet signal generator, model 7632, the output of which was attenuated by a factor of 100. Each amplifier was tested individually, for both pass band and gain. They were then checked by means of a square pulse generator with an output pulse of 0.08 microsecond duration and 0.05 microsecond rise time. It was fed through the three amplifiers, and each was checked for pulse shape and gain. The pulse length was doubled by the pre-amplifier, passed unchanged by the Hewlett Packard, and widened again by the linear amplifier. This widening was least (0.4 microseconds) for input time constant of 0.2 sec as expected, and for the intermediate feedback position; hence despite the lower amplification for this position, it was felt to be the best one to use. The gains of the pre-amplifier and linear amplifier were slightly less for pulses than for the sine wave signal from the generator. This was felt due to the pulse being faster than the amplifiers. As it was a fairly small difference ($<10\%$) and as the argon pulses were thought to be slower than those used here for calibration the gains were taken as those of the sine wave.

The high voltage supply for the argon chamber caused considerable trouble. A battery supply consisting of seven 300 volt Eveready Minimax batteries was used with the second chamber. The bakelite insulation used in it began to break down at 2100 volts negative, causing pulses to appear on the oscilloscope. These were at first roughly of the size expected for argon, and as they did not appear for positive fields, they were mistaken for argon pulses. Much work was done on them; however, after a while they began to increase in size, appear at the low voltages, and finally to appear
with positive fields. They were finally traced to the bakelite, which proved to be very faulty as insulation. Lucite was substituted for it and the spurious pulses disappeared. Later, when the pulses that were finally obtained were definitely proved to originate in the liquid argon, a Cintel stabilized power supply, type 1892, was substituted for the batteries. Its voltage could be more easily varied. The smoothing proved insufficient, and so an external RC filter was included.

It was found that the 2000 volts obtainable from it was frequently high enough to cause electrons to flow across the Kovar seal surface and appear as pulses. These were reduced if the glass surface were cleaned with ether, and were almost completely eliminated when the chamber was cooled. It was also found that a field of 80 kv per cm. caused dielectric breakdown of the argon between the plates; it was therefore necessary to work at fields of less than this value.

The input capacitances of the chamber and pre-amplifier were measured with a Q-meter. The latter had a value of 10.8 picofarads, while the former was 3.5 picofarads plus the capacitance due to the electrodes themselves. This varied from 0.5 to 1.2 picofarads depending on the electrode separation.

**Electrometer**

For the detection of a direct current from the pulses such as that done by Gerritsen (1948) and Hutchinson (1948) an electrometer of the type described by Dubridge and Brown (1933) was constructed. Its circuit is shown in fig. 7. The tube used is a Victoreen #5800 electrometer tetrode; the input grid resistors are the Victoreen HiMeg type with tolerances of < 1%. The tube was enclosed in a light metal cylinder which afterwards was
FIG. 7  ELECTROMETER CIRCUIT
filled with ceresin wax. This prevented the leads and glass from becoming contaminated with moisture and dirt, and eliminated any possibility of spurious photo-electric effects arising. The three ranges provided by the input resistors were connected into the circuit, as was the input lead, via small brass plugs. Each of these was surrounded by a cylinder of ceresin wax, one centimeter in radius and one centimeter long. This provided a very high resistance to ground ($\sim 10^{18}$ ohms) and negligible capacitance. A Tinsley type SS6/45 galvanometer, with a sensitivity of $6 \times 10^{-10}$ to $6 \times 10^{-9}$ amps/mm. was connected between the plate and first grid. In this way any variations in filament voltage or emission would affect each of these electrodes in a constant ratio, and thus not disturb the galvanometer reading.

To get the electrometer into working condition, the plate and first grid currents were set at their recommended values; the filament current, $I_f$, was varied to find the point at which the galvanometer deflection reached a maximum with respect to $I_f$. This was the optimum working point of the tube, but $I_f$ had then to be within 10% of its rated value of 10 milliamps. To achieve this, it was found necessary to increase the other two currents to well above their recommended values, viz. to 30 and 42 microamps for the plate and grid respectively. Calibration of the circuit was accomplished by applying a known voltage from a dry cell to a potential divider, and taking a fraction of it, about $10^{-3}$ volts, into the $10^8$ ohms input resistor. This gave a sensitivity for the instrument of $2.2 \times 10^{-13}$ amps/mm. for the galvanometer at its most sensitive setting. Thus it should be possible to observe a current of as low a value as $2 \times 10^{-15}$ amperes with the $10^{10}$ ohm input resistor.

When the electrometer was in operation, the zero was found to shift considerably with time, and required several hours to settle down. Furthermore grounding was very critical. Thus movement of any large objects,
so much as three feet away from the input lead, caused large deflections on the galvanometer, even for the least sensitive setting of the instrument. It was found impossible to completely shield the electrometer from this effect.

**ANALYSIS OF RESULTS**

**Pulses Obtained**

After considerable time had been spent on measurements of the spurious insulation breakdown pulses, their origin was finally discovered; they were eliminated, and no further pulses were discernible. Argon purification was attempted but failed to yield results. The next possibility was then investigated, viz., the pulses were below the noise level of the pre-amplifier. The latter was changed, its signal to noise level improved, and pulses were then observed. These were first found with the six tube pre-amplifier described previously on page 26, with it the signal to noise ratio was only about 2:1. With the third pre-amplifier used, whose circuit is given in fig 6(a), this ratio was improved to around 4 or 5 to 1.

The amplified pulses had the shape shown in figs. 8 (a) and 8 (b). These photographs were taken with an oscilloscope camera at 1/50 second exposure, with the oscilloscope sweep time set at 0.5 microsecond/cm. The pulses were obtained with fields of 33.3 kilovolts/cm. for fig. 8(a) and 16.7 kilovolts/cm. for 8(b), with the electrode spacing set at 0.03 cms. A few pictures were taken at 1/5 second exposure, to illustrate the pulse distribution, one of which is shown in fig. 8(c). Here E = 16.7 kilovolts/cm. and the scope sweep time was set at 0.2 microseconds/cm. The pulse sizes were given from the oscilloscope calibration figure and
FIG. 8(a)  ARGON PULSES 1/50 SEC. EXPOS.

FIG. 8(b)  ARGON PULSES 1/50 SEC. EXPOS.

FIG. 8(c)  ARGON PULSES 1/5 SEC. EXPOS.
the amplification of the pre-amplifier, the Hewlett Packard amplifier, and the Linear Amplifier. The oscilloscope sensitivity was set at 0.20 volts/cm for fig. 8(a) and 0.10 volts/cm for fig. 8(b) and 8(c); the total amplification was 490. These give sizes for the largest pulses observed on the two photographs of 450 microvolts and 310 microvolts for fields of 33.3 and 16.7 kilovolts per cm, respectively. The size of the pulses in fig. 8(b) had to be estimated roughly as the film used was insensitive to red light and so the scale did not appear. These values are slightly larger than those obtained by Davidson and Larsh (1950), whose maximum pulse sizes were 475 microvolts at 100 kilovolts per cm., and 270 microvolts at 29 kilovolts per cm., with an electrode spacing of 0.0094 cms.

Because there had been so much difficulty in obtaining pulses, and so many spurious effects observed, it was necessary to determine the origin of these pulses as carefully as possible. They did not occur when the field was reversed; This was important, but as mentioned before, breakdown will sometimes occur with only one direction of voltage, so it was not conclusive. The few small pulses occurring with a negative field are given in table #1 and graph #1, and are expected if the alpha range is of the same order as the electrode separation. These were also found by Davidson and Larsh but no quantitative data were given by them so that comparison is impossible.

There was no sign of pulses when the chamber was evacuated and cooled, provided that the kovar seal was perfectly clean. This eliminated the possibility of any spurious effects being caused by insulation breakdown. A further test proved that the pulses occurred in the liquid argon and then only when it was pure. Argon was liquefied in the chamber and pulses observed. The air trap was isolated from the chamber and cooled with liquid oxygen; oxygen gas was introduced into it under pressure and
liquefied. It was then connected to the chamber and the oxygen allowed to boil off and recondense in the chamber. Almost immediately the pulses observed in the argon disappeared entirely. Very little oxygen was needed to destroy them, but no quantitative measurements were made.

Finally, it is possible to attribute the pulses with certainty to ionization of argon by the alpha particles. The strength of the radioactive source is given by:

$$N_t = N_o e^{-\lambda t}$$

where $N_t$ = the number of particles observed per unit time at time $t$

$N_o$ = the number of particles observed per unit time at some arbitrary zero of time, ($t = 0$)

and $\lambda = 0.693 / T$ where $T$ is the half life of the element considered.

For radium F, the half life is 138 days. Three trials were made on the chamber for different values of the electrode spacing. These were taken 15 and 22 days after the first one, and gave values of $N_t$ of 4400 per min. at $t = 0$, 4060 per min. at $t = 15$ days and 3930 per min. at $t = 22$ days. According to the equation given above, $N_{15}$ and $N_{22}$ should be 4070 and 3920 respectively, values very close to the observed ones. Thus it can be concluded that the pulses observed and counted originate in pure liquid argon when an electric field is applied to it, and are due to the ionization of argon atoms by alpha particles.

An attempt was made to obtain pulses from the action of gamma rays on the liquid argon, similar to the method used by Davidson and Larsh. However, the chamber was not constructed for it, and could not be easily changed. Due to the much greater penetration of the gamma rays, it is
necessary to have a large gap (about 1 cm.) and to place the source about 2 cm. from the electrode centres. While the fields need not be as large as for alpha particles, they should be greater than 5 kilovolts/cm. to give observable pulses. The Kovar seal would not withstand the necessary voltages. However, a cobalt 60 source was placed in the liquid oxygen outside the chamber of which the gap was set at 0.1 cm, and a few pulses were observed with a negative field (as the alpha source remained in the system, it was necessary to use a negative field). Doubt resulting from the presence of the alpha source made this no more than suggestive of further work.

Rise Time Measurements

The difficulty in observing pulse shape is the obtaining of sufficient amplification over a wide frequency band. Due to the size of the pulses it is imperative to keep the noise level at less than 100 microvolts, zero-to-peak with 10^4 ohms input resistance of the pre-amplifier, but with a 10 Mc frequency band this is equivalent to a noise figure, F, of less than 2. Since the constant current pulses are converted by the input stage of the pre-amplifier to voltage pulses characteristic of charging up the input capacitance, the actual pulse durations are given by the rise times of the voltage pulses displayed on the oscilloscope. Some estimates of this rise time were made, using the 6 tube pre-amplifier and the Hewlett Packard amplifier. These gave rise time τ of 0.10 microseconds at E = 40 kilovolts/cm. and l = 0.05 cm., and 0.06 microseconds at E = 33.3 kilovolts per cm. and l = 0.03 cm. The corresponding mobilities are 12.5 cm^2/volt-second and 15 cm^2/volts-second respectively. These are slightly smaller than the values obtained by Malkin and Schultz (1951), which were 15 and 18 cm^2/volt-second for the two fields used above.

The mean free path L of the electrons in liquid argon is given by
the equation #3 on page 9, viz:

\[ L = \left( \frac{1.5 m M}{e^2} \right)^{1/2} K^2 E \]

\[ = 1.87 \times 10^{-13} K^2 E \]

for \( K \) and \( E \) in practical units. The values of \( L \) are then \( 1.18 \times 10^6 \) cms. and \( 1.38 \times 10^{-6} \) cms., giving an average of \( 1.3 \times 10^{-6} \) cms. From this is obtained a collision cross-section for the electrons with respect to the argon atoms. It is given by

\[ \sigma_{\text{coll}} = \frac{1}{L \rho} \]

where \( D \) is the number of argon atoms per cm.\(^3\). For liquid argon the density is \( 1.4 \) gms./cm.\(^3\) or \( 2.11 \times 10^{22} \) atoms per cm.\(^3\). Therefore, the collision cross-section is \( 3.7 \times 10^{-17} \) cms.\(^2\); of course this value is very rough, as it depends upon \( \tau^2 \); with an error of perhaps \( \pm 20\% \) in \( \tau \).

Two difficulties appear in the measurement of \( \tau \). One is the apparent inability of the oscilloscope used to trigger on the edge of the pulse. This is most noticeable on the faster sweep times. The other is due to the actual shape of the pulse. As the pulse top has become considerably rounded after passing through the amplifiers, the rise time determination is subject to large errors. If the capacitance is fairly large, \( \tau \) should be measured to the peak of the pulse, while if \( C \) is very small, the voltage will reach saturation, and \( \tau \) is measured to the point at which the pulse voltage begins to drop off. This difficulty is indicated in fig. 8(a) to (c), although of course these pulses have been shaped by the slower amplifiers used, and so their rise times are essentially meaningless.

The above method could possibly be improved by finding the optimum electrode spacing. The voltage is given by:

\[ V_{\text{peak}} = \frac{n e}{\tau} R \left( 1 - e^{-\tau/RC} \right) \]
and for $\tau \gg RC$, the exponential can be neglected, giving $V$ proportional to $1/\tau$. At the same time, as $\tau$ becomes small, the amplifiers must have a wider band width; there should be some value of $l$ and $E$ at which the signal to noise ratio is a maximum.

There are two other circuits which could possibly be used to find mobilities. One would employ a parallel inductance capacitance input; this will give a sinusoidal output for $t < \tau$, and a phase change at $t = \tau$ to a decaying sinusoidal wave of the same frequency. If this could be observed on an oscilloscope, and the phase change were large enough, $\tau$ could be found. The other circuit would differentiate the current pulse, producing two sharp blips of opposite sign, at time $\tau$ apart. If this signal could be amplified and fed into a circular delay line, and the latter observed at different points along it, a point could be found at which the two signals travelling in opposite directions around the line cancelled each other. From the position of this point $\tau$ could be found. However, the length of such a line would be too great to be easily handled.

Pulse Size Distributions

For the determination of pulse size distributions a narrower band of amplification is permissible, hence it is possible to get the noise down to about $1/4$ of the signal. Three trials were taken, using the circuit of fig. 6(b), with different settings of the electrode gap. The field was varied and a set of curves of normalized pulse number, $N/N_0$, against the size $V$ found for the different values of $E$, $N$ being the number of pulses of size $V$ or less. Table #1 and graph #1, taken for $l = 0.05$ cms., indicate the results obtained; the curves of $l = 0.067$ cms. and $l = 0.03$ cms. are similar in shape to these. Table #2 and graph #2 give the variation of $\Delta N/\Delta V$ with $V$, obtained from the curve for 40 kilovolts/cm. With
### TABLE 1  PULSE SIZE DISTRIBUTION FOR AN ELECTRODE SEPARATION OF 0.05 CMS.

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GRAPH 1. PULSE SIZE DISTRIBUTION FOR 0.05 cm ELECTRODE SPACING

PULSE SIZE (MICROVOLTS)

NOISE (MIN)^-1

NOISE 10 KV/CM 15 KV/CM 20 KV/CM 30 KV/CM 40 KV/CM

50 100 150 200 250 300 350 400 450

PULSE SIZE (MICROVOLTS)
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Graph 2. Pulse Size Frequency Curve for 40 KV/cm.
\[ \Delta V = 9 \text{ microvolts.} \]

The scaler accuracy was checked with the 60 cycle/second signal supplied in the instrument; it gave readings of well within 2% of the true value. Also it showed that a zero correction of 0.5 division was necessary, and all curves given here have been so corrected.

In calculations of the total number of pulses, \( N_t \), the maximum spread in the values of \( N \) used is about \( 5 - 10\% \); this is within the accepted experimental error. However, two effects do cause trouble in finding \( N_t \). These are noise at the small pulse size readings, and the very gradual approach of the high field curves to a steady value of \( N \) as the discrimination level is reduced. The former can be circumvented by discarding or correcting any readings taken at discriminator settings which fall below the maximum noise level. To eliminate the latter effect it is necessary to take only those values of \( N \) which definitely have reached a limit, in particular, those which follow some slight drop in the count. Taking such precautions it is possible to get a fairly accurate value of \( N_t \), e.g., 4060 counts per minute for the \( Z \approx 0.05 \text{ cms.} \) trial with a standard deviation of \( \pm 70 \) counts per minute.

As mentioned previously, three effects will cause a variation of \( N \) with the pulse size. The noise correction was the first to be made, and was accomplished as follows. The output of a pulse generator was fed into the linear amplifier, as shown in fig. 9, and the pre-amplifier connected to the chamber as before. The Hewlett Packard gain and the pulse generator output were varied to give a signal to noise ratio approximately the same as that found for a particular pulse distribution curve. Then, with the Hewlett Packard turned off, the distribution of pulses obtained from the generator was recorded. The Hewlett Packard was turned on, introducing the noise, and
FIG. 9  NOISE DETERMINATION CIRCUIT
the resulting distributions were obtained. These are given in table #3 and graph #3, for signal to noise ratios of 2:1; 3:1; 4:1; and 5:1. The pulse sizes are given as if they originated at the pre-amplifier input in order to compare them with the actual argon pulse distributions. It was assumed that the pulses from the generator were mono-energetic and any variation from a vertical line was due to scaler inaccuracy and the noise originating in the linear amplifier. Accordingly, the median pulse height found when no pre-amplifier noise was present was taken as their true height; the noise correction was simply the noise affected pulse size at any value of \( \frac{N}{N_t} \) minus this median value. Table #4 gives the result of this correction when applied to the 10, 20 and 40 kilovolt/cm. curves given in graph #1; the signal-to-noise ratio corrections applied are those of 2:1, 3:1 and 4:1 respectively. It can be seen from these tables that they fit quite well the actual ratios found for the argon pulses. The resultant curves are shown in graph #4. Also corrected were the 30 kilovolt/cm. curve for \( l = 0.067 \) cms. with signal to noise correction of 3:1, and the 16.7, 33.3 and 50 kilovolt/cm. curves for \( l = 0.03 \) cms., with signal to noise corrections of 3:1, 4:1 and 5:1 respectively.

According to the second cause of pulse size variation, viz., that due to a variation of transit time resulting from the angle of alpha emission, \( \frac{N}{N_t} \) should vary linearly with \( V \). A straight line was drawn to fit as well as possible the curves of graph #4. These lines as shown must fit the equation

\[
V = \frac{n e}{C} - \frac{n e}{C} \frac{R_0}{2l} \frac{N}{N_t}
\]

which gives

\[
V_{\text{MAX}} = \frac{n e}{C}
\]

\[
V_{\text{MIN}} = \frac{n e}{C} \left( 1 - \frac{R_0}{2l} \right)
\]
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GRAPH 3, NOISE CORRECTION CURVES

![Graph showing noise correction curves with pulse size in microvolts on the x-axis and signal-to-noise ratio on the y-axis. The graph illustrates the effect of pulse size on noise correction for different signal-to-noise ratios (2, 3, 4, 5).]
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GRAPH 4. NOISE CORRECTED PULSE SIZE DISTRIBUTIONS

\[ N/N_T \]

1.0

0.8

0.6

0.4

0.2

10 KV/CM.

20 KV/CM.

40 KV/CM.

PULSE SIZE (MICROVOLTS)
Thus \((V_{\text{max}} - V_{\text{min}})/V_{\text{max}}\) should be a constant with respect to the field and should equal \(R_{\alpha}/2\). The values of this for the straight lines shown in graph \#4, as well as for the other trials, are given in table \#5. The average range of the alpha particles is 0.012 cms, with a standard deviation of \(\pm 0.0015\) cms; this is larger than the value found by Davidson and Larsh (1950) by a factor of 2.4. The difference would be reduced if the weighted centre of ionization were considered to be further along the alpha track than \(R_{\alpha}/2\). This would be indicated by the ionization curves of alpha particles, and a value of about \(2/3\) \(R_{\alpha}\) would probably be closer to the centre of the average position of electron formation. This yields a value, \(R_{\alpha} = 0.009\) cms, still considerably above Davidson and Larsh’s (0.005 cms).

An attempt was made to determine whether the expansion to the first two terms only of the exponential in the equation giving \(V\) in terms of the input circuit components (equation \#2(a), page 15) was justified. Accordingly \(\exp\left(-\tau/RC\right)\) was approximated to by three terms while the expansion of \(\exp\left(-\tau_0/RC\right)\) was kept to two. This gives:

\[
V_\theta = \frac{ne}{C} R \left[ 1 - \left( 1 - \frac{\tau}{RC} + \frac{\tau^2}{2RC^2} \right) \left( 1 + \frac{\tau_0}{RC} \right) \right]
\]

which upon simplification and substitution for \(\tau_0\) is

\[
V_\theta = \frac{ne}{C} \left( 1 - \frac{\tau}{2RC} \right) - \frac{ne}{C} R_{\alpha} \frac{N_0}{N} \left( 1 - \frac{\tau}{RC} \right)
\]

with the term \(\tau^2/2RC^2\) neglected as small in comparison with the others. This gives for \(R_{\alpha}\) the following equation:

\[
\frac{R_{\alpha}}{2\ell} = \left( \frac{1 - \tau/2RC}{1 - \tau/RC} \right) \frac{V_{\text{max}} - V_{\text{min}}}{V_{\text{max}}} > \frac{V_{\text{max}} - V_{\text{min}}}{V_{\text{max}}}
\]

as \(\tau = l/KE\) and \(K \propto 1/\sqrt{E}\), \(\tau\) can be expressed as \(\tau = l/\alpha \sqrt{E}\) for an arbitrary constant. Hence the correction factor becomes

\[
\frac{1 - \frac{l}{\alpha RC \sqrt{E}}}{1 - \frac{l}{\alpha RC \sqrt{E}}}
\]
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This will \( l \to 0 \) or \( E \to \infty \). It will have the greatest effect on the curves obtained for large electrode spacings and small fields, and in all cases it will increase \( R_\alpha \).

As will be seen from table \#5 there is no systematic variation of \( R_\alpha \) with \( l \), and the slight difference occurring between the average value of \( R_\alpha \) for \( l = 0.05 \) cms. and that for \( l = 0.03 \) cms. would actually be increased by this correction due to its greater effect on values of \( R_\alpha \) obtained from the larger electrode spacing. There is a slight increase in \( R_\alpha \) with increasing field, which the above consideration would tend to correct. Using the value \( \tau = 10^{-7} \) seconds for \( E = 40 \) kilovolts/cm. and \( l = 0.05 \) cms., \( \tau/RC \) will be 0.067, while for 10 kilovolts/cm. \( \tau \) should be only twice as large, giving \( \tau/RC = 0.133 \); the correction terms will be 1.04 and 1.06 respectively. These are entirely insufficient to account for the difference of 0.002 cms. found, and as the correction factor is so much smaller than the standard deviation observed, that factor may be neglected. Therefore, for values of \( R \) and \( C \) used here, the exponential expansions may be limited to two terms with negligible error in calculating the alpha range in liquid argon.

The third cause of variation of \( N/N_T \) with pulse height is now considered. This is the effect that the angle of alpha emission will have on the number of electrons contributing to the current pulse, due to their recombination with positive argon ions. The equation derived for this effect, as given before (\#6, page 17), is

\[
m = n_0 \left( 1 - \frac{n_0 \tau^2}{\pi l R_\alpha} \sec \theta \right)
\]

This is assumed to hold only for \( \theta \leq 80^\circ \). The deviations of the distribution curves given by graph \#4 for \( N/N_T > 0.60 \) (i.e. for \( \sin \theta > 0.6 \)) were assumed to be due entirely to this variation of \( n \) with \( \sec \theta \); that is, the differences
between the observed pulse size and that given by the straight line for various values of $N/NT$ were subtracted from $V_{\text{max}}$. If the above equation were multiplied through by $e/c$, it would become

$$V \equiv \frac{n_o e}{c} = \frac{n_o e}{c} \left( 1 - \frac{n_o \sigma^2}{\pi \alpha R_\alpha} \sec \theta \right)$$

and the voltage values obtained above should fit this straight line, with $\sec \theta$ evaluated from $N/NT = \sin \theta$.

Table #6 and graph #5 give the variations obtained, for the two curves corresponding to $l = 0.05$ cms., $E = 40$ kilovolts/cm, and $l = 0.03$ cms., $E = 33.3$ kilovolts/cm. The curves are very roughly fitted by straight lines, with deviations from this line for both large and small values of $\sec \theta$. The levelling off of the curve for $\sec \theta > 3.5$ is expected, for the pulse size must approach a minimum value as $\sec \theta \to \infty$ (i.e. as $N/NT \to 1$). It seems to occur at values of $\theta$ of $\sim 65^\circ$, or $N/NT \sim 0.95$; this would indicate that the effective radius of the ion column is greater than $1/50$ of the alpha range. Variation from the straight line for small values of $\sec \theta$ could be due to inaccuracy in drawing the straight line representing variation of pulse size with $\sin \theta$. If this line were slightly steeper, the differences between it and the actual readings for $N/NT < 0.6$ could be made to fit the straight lines of graph #5. Such a change in slope would yield a smaller value of $R_\alpha$; however, this change would be < 10%, as the pulse size differences necessary to fit the points at $\sec \theta = 1.15$ and 1.00 to the straight line are quite small.

As the equation above contains three unknown quantities, viz., the constants $n_o$ and $r$, and $\sigma^2$ which would vary with field, no attempt was made to derive information from the slopes of the curves. Also, in order to determine either the radius of the ionization column or the capture cross-section, further information about one of them is necessary, as they cannot
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GRAPH 5. PULSE SIZE VARIATION WITH SEC θ

- ○ $1 = 0.05$ cm, $E = 40$ KV/CM
- × $1 = 0.03$ cm, $E = 33$ KV/CM
be isolated in the above equation. The number of electrons produced, \( n_0 \), can be isolated and estimated; inspection of the above equation shows that it would be equal to \((V_{\text{max}} + \text{slope})\ C/e\). This gives a value for \( n_0 \) which is not large enough to be independent of field, as it should be. According to the curve for \( l = 0.03 \text{ cms}, E = 50 \text{ kilovolts/cm.}, \) \( n \) is \( 5.5 \times 10^4 \), while \( n_0 \) found here, the maximum value of \( n \), is only \( 4.5 \times 10^4 \). As the variation in pulse size due to recombination with argon ions is independent of the electrode separation, and the field should have no effect on the number of electrons produced, \( n_0 \) should not vary with either \( l \) or \( E \). No explanation of this discrepancy has been found.

Variation of Maximum Pulse Size with Field

As shown in table #5, there is considerable variation in the maximum pulse size obtained for various settings of the field and electrode separation. A relationship between this pulse size and the field was sought, and from table #7 and graph #6, it seems that the data fit quite well on a straight line giving maximum pulse size proportional to \( E^{1/2} \). There are two possible origins of such a variation: one is due to the electron transit time, \( \tau \); the other is due to the number of electrons contributing to a pulse, \( n \).

If, in the equation

\[
V = \frac{ne}{\tau} R \left(1 - e^{-\tau/RC}\right)
\]

\( \tau \) is greater than \( RC \), the exponential term is small and may be neglected. This would give \( V \propto l/\tau \), or since \( \tau \propto l/\sqrt{E} \) as shown before, the pulse size will vary inversely with \( l \) and directly as \( \sqrt{E} \). The relationship given by graph #6 would seem at first to indicate this. However, the condition that \( \tau \) be large with respect to \( RC \) is directly contrary to the assumption made in deriving the pulse distribution variation with \( \sin \theta \) (which variation
<table>
<thead>
<tr>
<th>l (cm)</th>
<th>E (KV/cm)</th>
<th>E (V/cm)^1/2</th>
<th>V_{max} (µV)</th>
<th>Ln(1-V_{max}/V_0)</th>
</tr>
</thead>
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<tr>
<td>.05</td>
<td>10</td>
<td>100</td>
<td>200</td>
<td>-.10</td>
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<td>440</td>
<td>-.24</td>
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<tr>
<td>.03</td>
<td>50</td>
<td>224</td>
<td>570</td>
<td>-.32</td>
</tr>
</tbody>
</table>
GRAPH 6. MAXIMUM PULSE SIZE VARIATION WITH FIELD

PULSE SIZE (µV)

0 100 200 300 400 500

E^{1/2} (VOLT/CM.)^{1/2}

○ l = 0.05 cm
× l = 0.03 cm
would not exist if $\tau \gg \tau_C).$ Also, it would demand that $\tau$ be larger than 1.5 microseconds - a factor of 10 greater than any transit time so far measured for liquid argon, with electrode spacings $\sim 0.05$ cms. Hence the variation in $V$ with $E$ cannot be explained on the basis of dependence of $F$ upon the transit time.

The second possibility is that, with the pulse size given by

$$V_{\text{max}} = \frac{ne}{C}$$

as stated previously, $n$ will be a function of field, due to the recombination of electrons with the argon ions being more frequent at low fields. This would necessitate $n$ increasing with $\sqrt{E}$, not linearly, but approaching a saturation value of $n_0$, at which no recombination will occur and all the free electrons created by the alpha particle contributing to the pulse. If this is the case, which seems much more likely than the first mentioned cause, the curves given in graph #6 represent only short segments of the curve, for which no curvature is observable. Under such circumstances all attempts to obtain $n_0$ from the data is impossible; much larger fields would be necessary, so that $V_{\text{max}}$ could be observed to level off and approach a value corresponding to $n = n_0$.

Davidson and Iersh (1950) used the equation

$$V_{\text{max}} = V_0 \left( 1 - e^{-\frac{a E}{n}} \right)$$

where $a$ is an arbitrary constant and $V_0$ was taken as the observed pulse size in argon gas, in which very little recombination would occur. The linear relationship of graph #6 would follow from this if $\sqrt{E}$ were $\ll 1/a$ for all fields used here. This equation can be changed to

$$\ln \left( 1 - \frac{V_{\text{max}}}{V_0} \right) = -a \sqrt{E}$$

and $V_0$ can be calculated on the assumption that $\rho/x$ is 25.4 eV, as it is
in argon gas. \( n_0 \) is then equal to \( 2 \times 10^5 \) electrons and \( \text{Vo} \) to 2100 microvolts.

A plot of \( \ln \left(1 - \frac{V_{\text{max}}}{\text{Vo}}\right) \) as given in table #7 against \( \sqrt{E} \) yields a straight line (graph #7). There is a slight concavity toward low values of the logarithmic variable for the \( l = 0.05 \) cm. curve. It would be caused by \( 1 - \frac{V_{\text{max}}}{\text{Vo}} \) being too low, i.e., by \( \text{Vo} \) being too small. This indicates the possibility of \( \frac{\rho}{x} (=\frac{\text{H}}{\text{n}_0}) \) being smaller than 25.4 eV., giving a conduction band for liquid argon. However, no such concavity can be found on the curve for \( l = 0.03 \) cms., so the issue remains in doubt.

The two curves obtained for different values of \( l \), and their slightly different slopes can be explained by the correction term derived in the previous section. It gives for the maximum pulse size obtainable for particular values of spacing and field, the equation

\[
V_{\text{max}} = \frac{n_e}{C} \left(1 - \frac{R}{RC}\right)
\]

As the last term in the expression has a value of only 0.07 for \( l = 0.05 \) cms. and \( E = 40 \) kilovolts/cm., as found before, it will not be sufficient to account for the difference between the two curves of graph #6 (\( \sim 15\% \)) but it will decrease this. A further possibility is that part of this difference is caused by capture of electrons by impurity atoms. Unlike recombination effects, the number of electrons removed by the capture process will vary directly with the electrode spacing. The magnitude of this process is unknown but the purification preformed here had no observable effect on it.

**Ionization Current Measurements**

In order to determine the ionization current, the electrometer was connected directly to the chamber, and allowed several hours to reach equilibrium. The chamber was cooled, argon introduced and the field turned on, as before. The deflections obtained were very large (\( \sim 10^{-11} \) amps.), while the actual currents calculated from a value of \( n_0 \) of \( 2 \times 10^4 \) electrons
GRAPH 7. MAXIMUM PULSE SIZE VARIATION WITH FIELD

\( \ln(1 - \frac{V_{\text{max}}}{V_0}) \)

-1.00
-1.50
-2.00
-2.50
-3.00

\( E^{1/2} \) (VOLT/CM.\(^{1/2}\))

100 125 150 175 200 225

○ \( 1 = 0.05 \text{ cm.s.} \)

× \( 1 = 0.03 \text{ cm.s.} \)
per pulse with 60 pulses a second should have been about $10^{-13}$ amps. These large deflections were accompanied at high fields with fluctuations of the same order occurring at a frequency of about one per second or so. These fluctuations persisted when the argon was removed. Their size was lessened considerably by removal of the external filtering supply on the high voltage, and by thorough cleaning of the external surface of the kovar seal. They could not be eliminated, and while they did not appear at the lower voltages, the steady deflection did, and was itself so much larger than the currents originating between the electrodes that it rendered the latter impossible to ascertain. There is a strong possibility that this steady current was due to tracking currents across the surface of the kovar seal, both inside and outside the chamber; like so many such effects it did not occur with equal magnitudes for both directions of the field, being larger when the chamber was positive with respect to the connection through the seal.

To overcome this, a large kovar seal is indicated for such use, with the gap between the electrodes kept small to enable the utilization of lower voltages to obtain the fields desired. Also more careful construction of the input circuit of the electrometer would probably be advantageous, with the input resistors completely shielded along with the tube itself. However, the most important change would be the introduction into the chamber of a much stronger alpha source. This was not done, as one yielding 50 - 100 counts per second was considered best for rise time and amplitude measurements. Where only direct currents need be measured, sources of 100 - 1000 times this activity would be permissible. With such a source (about 5000 counts/sec.) the ionization current should be greater than $10^{-11}$ amps for fields of both positive and negative values. (The positive ions will naturally yield as large a current in moving across the gap as the electrons do, although, with their much lower mobilities, the pulses them-
selves are not discernible.) With such a source it should be possible to determine the ionization currents corresponding to different values of electrode spacing and field, and thus achieve an independent method of calculating n.

**DISCUSSION**

From the attempts to determine both the transit time, $\tau$, and the pulse amplitude, using the same circuit, it must be concluded that such an approach is essentially impossible. The amount of information available from the voltage pulse, (resulting from a steady current, of magnitude $ne/\tau$ and duration $\tau$, when it passes through an RC parallel circuit) is limited, and due to the values of n and $\tau$ is too small to yield significant results for both quantities. Some sacrifice of either transit time or amplitude accuracy is essential if the other quantity is to be accurately measured. As amplifiers with good signal to noise ratios were available, it was decided to pursue pulse size distribution data and neglect transit time measurements, which demand much faster amplifying equipment.

The mean free path and collision cross-section of an electron in liquid argon were estimated from the transit time for two values of field and electrode spacing. The cross-section was found to be $4 \times 10^{-17}$ cms.$^2$; this compares well with the value obtained by Malkin and Schultz (1951) but differs considerably, as does their value, from those previously published for argon gas (Brode 1933); these give a maximum probability of capture for the electron energy values used here, due to the Ramsauer effect.

The observed magnitude of any pulse at a time $t$ after its start has been shown to be given by
\[ \mathcal{V} = \frac{ne}{t} R \left( 1 - e^{-t/R} \right) \]

for \( 0 \leq t \leq \mathcal{T} - t_0 \). This pulse size reaches a maximum at \( t = \mathcal{T} - t_0 \) which will not be the same for all pulses, due to variation of \( \mathcal{V} \) with several quantities not explicitly entering the above equation. The noise of the amplifying components produces fluctuations in pulse size as shown in table #3 and graph #3. The quantity \( t_0 \) varies with the angle of alpha emission, \( \Theta \), as does the number of pulses originating from alpha particles emitted at angles \( \Phi \). Furthermore the number of electrons contributing to a pulse will vary with sec \( \Theta \) and with the applied field, due to recombination. There will also be second order effects, caused by \( \mathcal{T} \) being proportional to the electrode spacing, \( l \), and inversely proportional to the square root of the field. Finally there will be some variation in pulse size due to the alpha particles not being completely mono-energetic, and to electron trapping by impurities. All these effects have been considered in the analysis of results, and the possible conclusions made.

The alpha range, \( R_\alpha \), has been found to be 0.012 cms., or, corrected for the heavier ionization occurring near its limit, to 0.009 cms. Both values are considerably larger than that suggested by Davidson and Larsh (1950), viz., 0.005 cms. In either case, the weighted centre of the electron column can be given a 0.006 cms. along the track of the alpha particles.

The number of electrons contributing to a pulse was found to vary between \( 2 \times 10^4 \) and \( 6 \times 10^4 \) depending on the field. It shows no sign of saturation at large fields, and could be as high as \( 2 \times 10^5 \) without a conduction band being present. Thus there is no reason as yet to demand conduction bands for liquid argon; higher fields would be necessary to determine this.
Finally, from the attempts to measure ionization current, it is apparent that more care is necessary in the construction and operation of the chamber and the electrometer circuit. Also a much stronger alpha source would be a definite advantage.
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