THE IONIZATION OF LIQUID ARGON BY α -PARTICLES

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Summary

Calculations suggest that in liquid argon the column of ionization produced by an α -particle consists of a core of positive charge $0.6-1.5 \times 10^{-5}$ cm in diameter concentric with a larger cloud of electrons extending to a diameter of 40×10^{-5} cm. This leads to a new description of charge separation in an applied electric field, in which the positive and negative charges are drawn apart slightly (polarized) and the more energetic electrons escape from the edge of the column. The remaining electron cloud reaches a state of dynamic equilibrium with the excess positive charge in a time short compared with that required for ionic recombination, which does not have a significant effect on the ionization current. The distribution of electron energies in the column of ionization is calculated from the results of recent ionization current measurements, and indicates that a 5.3 MeV α -particle expends 0.5 MeV as kinetic energy of the liberated electrons.

I. INTRODUCTION

Measurements of ionization currents in α -irradiated liquid rare gases (Williams and Stacey 1957) showed that the currents were too large to be explained by either of the existing theories of ionic recombination (Jaffé 1913; Kramers 1952). Both of these theories assume that the electrons released by ionization form heavy negative ions by attachment to neutral atoms, immediately they are formed. This is a questionable assumption for any material, but in liquid argon it is quite certain that the electrons remain free altogether (Williams 1957). Liquid argon is therefore probably the simplest material to re-examine theoretically.

The theories of Jaffé (1913) and Kramers (1952) describe columns of positive and negative ions which have similar mobilities and which, in the absence of an applied field, remain completely superimposed. The application of a field to such a system was considered to draw the two opposite charge clouds apart at a rate dependent upon the field, while ionic recombination removed ions of both signs from those parts of the charge clouds which overlapped. The increase of ionization current with field was thus explained as the reduced opportunity for recombination when the two charge clouds were drawn apart more rapidly, and the general features of the ionization current versus field curve were explained for a number of materials.

Gerritsen (1945), who experimented with a number of liquids, including argon, and who was responsible for the posthumous publication of Kramers' theory, maintained that this theory fitted his data for liquid argon reasonably well. It appears, however, that neglect of the mutual attraction of the opposite

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charge clouds makes the theory quite invalid in the case of liquid argon, and possibly for most other materials.

Considering only the charge collected from a single α -particle track in a small field and neglecting the charge which is lost in ionic recombination, we see from the experimental data (Williams and Stacey 1957) that about 7 ion pairs are collected per volt/cm of applied field; i.e. in a field of F V/cm, = V e.s.u., a total charge of (7eF) is collected per α -particle. The charges are drawn apart with a force (7eF)V, but at a separation d, significantly larger than the column diameters, they experience a mutually attractive force $2(7eF)^2/\varepsilon ld$, where l is the length of the columns and ε is the dielectric constant of the liquid. Thus for the applied field to succeed in drawing the charges apart

 \mathbf{or}

 $7eFV > 2(7eF)^2/\varepsilon ld$,

$d > (14e/\epsilon l) \cdot F/V = 2 \cdot 8 \times 10^{-4} \text{ cm.}$

The charge clouds were considered to have diameters less than one-tenth of this value, so that the applied field only competes favourably with the mutually attractive force when the charge clouds are already well separated. This indicates that the observed charge collection cannot occur by the Jaffé-Kramers process.

In the theory considered here the kinetic energy imparted to the electrons by the ionization process plays an important part in overcoming their strong attraction to the positive ion column. The fraction of the α -particle energy which goes into kinetic energy of the electrons is estimated in Section IV. The electrons lose their kinetic energy only after many collisions, of the order 10^5 in argon (a discussion of this point is given by Williams (1957)) so that they form a diffuse cloud of greater diameter than the positive ion column.

The electron cloud remains in dynamic equilibrium in the central field of the positive charge, while it is gradually reduced by recombination in the central region and loss of energy by collision of electrons with atoms.

In an external field perpendicular to the column of ionization the centres of the positive and negative charge clouds are separated or polarized by an amount which is calculated in Section III. This enhances the probability of escape from the column of the more energetic electrons, which during their random motion have sufficient energy to reach the critical radius at which the applied field becomes greater than the central field.

The motion of the electrons is such that, if a particular electron has sufficient energy, it will escape from the column merely because by chance it will eventually cross the critical radius in a direction favouring escape. The remaining less energetic electrons are more tightly held by the excess positive charge of the column, so that, however long the column lasts before recombination destroys it, it can make no further contribution to the ionization current but drifts bodily to the negative electrode.

A rigorous analysis of the model presented is virtually impossible since it depends essentially upon the initial distribution of electron energies, itself only

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obtainable by numerical methods (e.g. Erskine 1954). This distribution is further complicated by the fact that the fastest electrons cause secondary or feather ionization, in tracks of low density, thus producing slow electrons well clear of the main column of ionization.

However, the main features of the theory are examined quantitatively in the following sections.

II. STRUCTURE OF AN ION COLUMN

The mean free path of positive ions is short, and collisions of heavy ions with neutral atoms rapidly reduce the kinetic energy of the ions to thermal energy. With respect to electrons, which are slowed down only after many collisions, the positive ions are thus considered to remain in a stationary column. The kinetic energy of the electrons then enables them to expand to a cloud of larger diameter against the potential energy of attraction to the positive ions. The density distribution of electrons in the ion column is not needed for approximate calculations, but the potential energy of a simple distribution can be calculated as a demonstration of the magnitude of the electron cloud expansion which is possible with a moderate electron kinetic energy.

The field F, at a distance r from the centre of any cylindrically symmetrical charge distribution of infinite length is due to the charge per unit length, ne/l inside a radius r, and has the value

where ε is the dielectric constant of the medium. Charge outside r does not contribute to the field at r. An electron in the ion column which is "expanding" past the radius r experiences a retarding field according to equation (1), where (ne) is the net positive charge inside r and l is the length of the column, supposing it to be much greater than the diameter. If r is outside the positive ion column, then n is also the number of electrons which have already expanded past r.

For the purpose of making an estimate of the potential energy of an "expanded" electron cloud for comparison with the kinetic energy available to the electrons, we may consider an ordered collapse of a uniformly dense cylinder, radius R, of N electrons into a smaller uniform cylinder radius R_0 of positive ions, exerting a field as in equation (1). The electrons inside R_0 may be considered as neutralizing (NR_0^2/R^2) positive ions, leaving $N(1-R_0^2/R^2)$ positive ions inside R_0 and the same number of electrons outside. The collapse occurs by electrons crossing R_0 , beginning with the innermost shell, and neutralizing the first available positive ions.

An elementary cylinder of electrons, thickness du, collapsing from a radius u, experiences a central field

$$F = \frac{2Ne(1-u^2/R^2)}{\epsilon lr}$$
 at radius r.

It reaches the first un-neutralized positive ions at radius v given by

$$u^2/R^2 = 1 + v^2(1/R^2 - 1/R_0^2)$$

The number of electrons starting from the element du is $N \cdot 2udu/R^2$, so that the energy of collapse of electrons from this element is

$$\mathrm{d} E = \frac{4N^2 e^2 u (1 - u^2/R^2) \mathrm{d} u}{\varepsilon l R^2} \int_v^u \frac{\mathrm{d} r}{r} = \frac{4N^2 e^2}{\varepsilon l R^2} u \left(1 - \frac{u^2}{R^2}\right) \ln \left(\frac{u (1/R_0^2 - 1/R^2)^{\frac{1}{2}}}{(1 - u^2/R^2)^{\frac{1}{2}}}\right) \mathrm{d} u.$$

The total energy of collapse is found by integrating with respect to u between R_0 and R:

$$E = (N^2 e^2 / \varepsilon l) \{ \ln (R/R_0) - \frac{1}{2} + \frac{1}{2} (R_0/R)^2 \}.$$
 (2)

Numerical solution of equation (2) shows that if the electrons reach a total potential energy of 0.5 MeV, they could expand to a uniform cylinder of radius R approximately three times the radius R_0 of the positive ion column.

This result can serve only as an order of magnitude because it is not imagined that uniform distributions describe the actual state of the ion columns.

III. POLARIZATION OF THE ION COLUMN

If the electron density in the central region is uniform the polarization or separation of the centres of the positive ion and electron distributions by an applied field is readily calculated. The density of electrons is defined by the characteristic radius R, the radius of the cylinder which the electrons would occupy if the whole distribution were of the same density as the central region.



Fig. 1.—Calculation of the attractive force between overlapping cylinders of positive and negative charge.

Consider the force of attraction of an elementary hollow cylinder of dN positive ions, with radius R_0 and centre Q, to the centre P of a uniform cylindrical cloud of electrons, at a distance d from Q as in Figure 1. The component in the direction PQ of the force on an element $d\theta$ at θ is

$$\mathrm{d}f = \frac{2N(r_1^2/R^2)e \cdot \mathrm{d}Ne(\mathrm{d}\theta/2\pi)}{\varepsilon lr_1} \cos \varphi,$$

since only the electrons inside radius r_1 are effective, where

$$r_1 \cos \varphi = d - R_0 \cos \theta.$$

Integrating over θ :

$$f = \frac{2Ne^2d \cdot \mathrm{d}N}{\varepsilon lR^2}.$$

Since this is independent of R_0 , the force on a solid cylinder of N positive ions with any radial distribution is $2N^2e^2d/\epsilon lR^2$.

This is equated to the force of the applied field, F, tending to pull the electrons away from the positive ions. Assuming n of the N electrons to have escaped from the column, this is

$$F(N-n)e=2N^2e^2d/\varepsilon lR^2,$$

$$d=\varepsilon lR^2F(N-n)/2N^2e. \qquad (3)$$

The parameter R is a function of n, since, by virtue of the escape of n electrons, the density in the central region is reduced. However, the fastest electrons would have spent only a small fraction of the time in the central region and not very greatly added to the density; also the net positive charge remaining tends to reduce the size of the electron cloud slightly, so that the overall error which results from taking R as constant is not great.

IV. ENERGY SPECTRUM OF THE ELECTRONS

The process of expansion of the electron cloud is similar to diffusion; each electron may traverse the positive ion column several times before the equilibrium state is reached. At equilibrium each electron conserves its total (kinetic plus potential) energy, the potential energy being gained by motion outwards against the central field described by equation (1), in which n is a function of r, determined by the electron distribution. During the expansion this distribution is time-dependent, so that any attempt to derive it from a given initial distribution of electron kinetic energies would be difficult, and this would be a necessary preliminary to determining the loss of kinetic energy during expansion and the ionization current. Here a much simpler calculation of the electron energy spectrum from measured ionization currents is made.

If the *n* most energetic electrons escape from the column in a field F perpendicular to the column, then the remaining cloud of (N-n) electrons has a critical outer radius r_c at which the total field is zero in the direction of F. The (n+1)th most energetic electron does not quite have enough initial kinetic energy to reach r_c .

Allowing for polarization as in Section III, we have for the field at r_c

$$0 = \frac{2Ne}{\varepsilon l(r_c + d)} - \frac{2(N - n)e}{\varepsilon lr_c} - F,$$

where the first term is attraction to the N positive ions, the second term is repulsion by the (N-n) remaining electrons, and the third term is the applied field. This gives

$$r_{c} = \left[\frac{ne}{\varepsilon lF} - \frac{\varepsilon lF(N-n)}{4N^{2}e}R^{2}\right] + \sqrt{\left\{\left[\begin{array}{c}\right]^{2} - \frac{(N-n)^{2}}{N^{2}}R^{2}\right\}}. \quad \dots \quad (4)$$

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A maximum value of R is set by the conditions that r_c must be real at all values of F. Measurements at the highest field used by Williams and Stacey (1957) give

$$R < 0.9 \times 10^{-5}$$
 cm.

In the following numerical calculations the value $R=0.75\times10^{-5}$ cm is taken; this is negligible at all but the highest fields and indicates that polarization is only important at high fields.

CALCULATION OF SPECTRUM OF ELECTRON ENERGIES								
F (V/cm)	n	r_{c} (10 ⁻⁵ cm)	ΔE (eV)	<i>E—E′</i> (eV)	$-\frac{1}{N}\frac{\mathrm{d}n}{\mathrm{d}E}$			
1	4.90	$19 \cdot 3$	0.404 × 10-8	$5\cdot 29_{48}$	0.139			
$3 \cdot 162$	18.6	23.3	1.41×10^{-3}	$5\cdot 29_{43}$	0 · 139			
10	57.6	$22 \cdot 8$	5.20×10^{-3}	$5 \cdot 29_{29}$	0.138			
$31 \cdot 62$	$2 \cdot 04 \times 10^{2}$	$25 \cdot 5$	9.29×10^{-2}	5·2876	0.139			
100	$7\cdot25 imes10^2$	28.6		$5\cdot 26_{55}$	0.109			
316.2	$1\cdot70 imes10^3$	$21 \cdot 3$	4.80 × 10 -	$5 \cdot 21_{69}$	0.095			
103	$3\cdot99 imes10^3$	$15 \cdot 7$	0.120	$5 \cdot 09_{06}$	0.0881			
$3\cdot 162 imes 10^3$	$8\cdot 32 imes 10^3$	$10 \cdot 4$	0.405	$4 \cdot 83_{24}$	0.0812			
104	$1\cdot 68 imes 10^4$	6.56	0.075	$4 \cdot 34_{73}$	0.0751			
$3\cdot162 imes10^4$	$3\cdot 02 imes 10^4$	3 ∙65	0.955	3 · 39 ₂	0.0670			
105	$5\cdot 31 imes 10^4$	1.89	1.64] • 75 ₃	0.0721			
$3\cdot 162 imes 10^5$	$9\cdot12 imes10^4$	0.815	1.75	0	0.1772			
					1			

		TABLE	1	,	
CALCULATION	OF	SPECTRUM	OF	ELECTRON	ENERGIES

If *n* electrons reach r_c in a field *F* and (n+dn) electrons reach (r_c+dr_c) in a field (F+dF) then the energy difference dE between the *n*th and (n+dn)th most energetic electrons is

$$dE = (2ne^2/\varepsilon lr_c)dr_c - \{(F+dF)(r_c+dr_c-r_0)e - F(r_c-r_0)e\},\$$

$$dE/dn = (2ne^2/\varepsilon lr_c - eF)dr_c/dn - (r_c-r_0)edF/dn,\qquad(5)$$

where r_0 is the effective radius from which the electrons have expanded, for which the r.m.s. radius of the positive ion column is taken. To obtain an approximate value of r_0 we may put $r_0 = R_0/\sqrt{2}$, where R_0 is the characteristic radius of the positive ion column. Adopting the result of Section II, $R_0 = \frac{1}{3}R$, we have

$$r_0 = R/3\sqrt{2} \simeq 0.2 \times 10^{-5}$$
 cm.

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In equation (5) n is the number of electrons with energy E or greater, equated to the number of electrons escaping to give the observed ionization current in an applied field F.

Values of r_c were calculated from equation (4), using the results of Williams and Stacey (1957). These were used to find dr_c/dn graphically and hence dE/dn, from which the spectrum of electron energies shown in Figure 2 was obtained by numerical integration. Details of the calculation are given in Table 1. E' represents an arbitrary energy because the lower end of the spectrum is experimentally inaccessible, but a reasonable estimate of E' can be made from the fact that the area under the curve must integrate to unity.



Fig. 2.—Energy spectrum of electrons in α -ionized liquid argon.

Drawing the spectrum with a sharp cut-off at $(E-E')=5\cdot 3$ eV is necessitated by the fact that the experiments extended down to the collection of 5 electrons per α -particle, so that the area under the curve beyond the last experimental point is almost invisibly small.

The peak extending from $5 \cdot 3$ eV downwards is evidently a result of secondary ionization, by which the very energetic electrons produce secondaries well clear of the main column of ionization. In terms of the present theory the secondary electrons are produced with appreciable potential energy. The spectrum of Figure 2 thus represents final energies of electrons in the column, being a modification of the spectrum of initial kinetic energies. The sharpness of the cut-off at $(E-E')=5\cdot 3$ eV is evidently due to an excitation of argon atoms at this energy, so that more energetic electrons rapidly lose their excess energy.

The cut-off is clearly at a lower energy than the first ionization energy of argon at 16 eV. This would require the curve of Figure 2 to double back to low values of (1/N)dn/dE at low E in order that the area under the curve shall integrate to unity, and then the total energy represented by the spectrum would be at least 2 MeV. Since this would allow no energy for excitations, double ionization, and positive ion kinetic energy, it is clearly inadmissible, although the results of Ward (1954) indicate that the energy lost by excitation is small.

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More reasonable is the simple extrapolation of the curve to the point at which the area under it is unity. This indicates $E' \sim 2 \text{ eV}$, in which case the total energy in the spectrum is approximately 0.5 MeV. This is the result used in Section II.

V. DISCUSSION

In the preceding sections a number of assumptions have been made, requiring more detailed consideration.

Ionic recombination has been ignored, but its importance depends upon the time scale of the processes described. However, the mean free path for scattering of the electrons can be calculated from the electron mobility data of Williams (1957). It varies with energy, being of the order 10^{-5} cm, so that an electron would be expected to experience an average of one scattering collision per traversal of the positive ion column. The density of positive ions in the column averages less than 10^{-5} of the neutral atom density, so that encounters with positive ions are of the order 10^{-5} times as frequent. The electrons would therefore need to make some 10^4 traversals of the positive ion column during the expansion for recombination to have a noticeable effect on ionization current, and the relatively long mean free path clearly indicates that this is highly improbable.

That the electron cloud should retain a cylindrical form in an applied field is a simplifying assumption. The cloud will be misshapen but the polarization considered in Section III gives an approximation to the field experienced by an escaping electron. The fact that the polarization effect is negligible except in high fields suggests that only at high fields (low electron energies) could the results of the present calculation be in error on account of distortion of the electron cloud.

In deriving equation (5) it was assumed that the energy expended by an electron in moving outwards against the central field of the positive ions is independent of the applied field. The correctness of this assumption depends upon the mechanism of electron cloud expansion. At low fields an electron of average energy may be partially screened from the positive ions by more energetic electrons, which are temporarily at a smaller radius, but which would escape from the column at an early stage in the expansion in a high field. The magnitude of this effect is not known, so that in equation (5) it is assumed that the expansion of electrons to any particular potential energy has the same form independent of the applied field.

Experimental data are not available for ionization currents due to α -particles from collimated sources. These would be necessary for correct application to the theory presented which concerns only the escape of electrons in fields perpendicular to the α -particle tracks. Jaffé (1913) recognized this problem, but merely stated that the fields considered in his theory were the components of the applied fields perpendicular to the particle tracks, i.e. that escape from the ends of the columns of ionization could be neglected. This was reasonable in the Jaffé-Kramers theories, since the probability of recombination during the drift of an ion through a large fraction of the ions of opposite sign was almost

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unity. In the present theory, recombination is much less probable since the positive ions and electrons have very little overlap and therefore escape of electrons in a field parallel to the column cannot be neglected altogether. The pulse experiments of Williams and Stacey (1957) indicate that the direction of α -particle emission with respect to the applied field does influence ionic collection, although the effect is smaller than that of Jaffé's theory. This effect is not considered in the foregoing theory, which therefore uses a "smoothed out" ionization current curve. Its influence on the calculations may introduce an error up to 50 per cent. in the estimated radii of the column and the energy spectrum.

The estimate of r_0 which was made in Section IV must be regarded as a lower limit, since it was obtained by considering the characteristic radius R of the central region of the electron cloud to enclose all the electrons, whereas it merely reflects the density of the central region. The upper limit is set by $r_0 = R = 0.75 \times 10^{-5}$ cm. This range of values puts an additional 20 per cent. possible error on to the value of -(1/N)dn/dE at (E-E')=0 in Figure 2, in which the upper limit is indicated.

The values of n and r_c in Table 1 do not give the electron density at any radius in zero field, since r_c has been calculated as the radius which n electrons can reach with the assistance of an external field. In the absence of an external field, the extreme radius of the column is approximately 20×10^{-5} cm.

A primary electron energy spectrum may be inferred from the results of calculations by Erskine (1954), who plotted the cross section Q for ionization of helium atoms as a function of the wave number k of the liberated electrons. The distribution may be approximately represented by a simple analytical expression

$$Q = Ck \exp\left(-\frac{k^2}{\sigma^2}\right),$$

where C and σ are constants. This leads directly to a spectrum of primary electron energies :

$$\frac{\mathrm{d}N}{N} = \frac{1}{E_0} \exp\left(-\frac{E}{E_0}\right) \mathrm{d}E,$$

in which E_0 is the average energy. When modified by a cut-off energy and secondary maximum as discussed in Section IV, this is not inconsistent with the appearance of Figure 1, but the energy range indicated by Erskine (wave numbers of the order 10^8 cm⁻¹) is clearly too large, giving an average electron energy in excess of 100 eV.

Liquids or gases with positive electron affinities evidently still require the Jaffé-Kramers treatment, although a modification of the assumption of identical positive and negative ion distributions must be strongly suggested. A large diameter of the negative ion cloud would result if the electrons began the expansion process as in argon before becoming attached to neutral atoms. This allows a larger ionization current than the original Jaffé-Kramers theories and avoids the objection to these theories which is raised in Section I.

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