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Ionization of liquid argon by α particles

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Abstract. From an analysis of electron pulses in liquid argon produced by α particles the average energy required to form an ion pair is evaluated, assuming Jaffé's theory of columnar recombination is valid. The mean value of 26·0 ev found in this way agrees well with the corresponding value of 26·4 ev for α particles in argon gas. Application of Jaffé's theory to the liquid state is discussed, and from the pulse distributions it is established that the experimental results are in good agreement with this theory.

1. Introduction

A considerable body of information has been accumulated dealing with the electrical properties of liquid argon. In particular, the mobility of electrons has received special attention because of an interesting property of this particular liquid which enables the electrons to remain as free entities of high velocity, rather than attaching to the liquid atoms to form massive ions (Williams 1957, Swan 1964). The influence of impurities on the electronic mobility has also been studied, and the attachment coefficient for oxygen when present in dilute solution has been measured (Swan 1963). The most outstanding feature of the results from these investigations is the similarity between corresponding measurements in liquid argon and in gaseous argon. In fact, the behaviour of electrons in liquid argon containing small quantities of molecular impurity gases can be most simply interpreted in terms of gaseous kinetic theory, as indeed can the attachment coefficients for oxygen. It is the purpose of this present paper to evaluate the energy required to form an ion pair during ionization of liquid argon by α particles and to compare this with the corresponding value obtained in gaseous argon, and also to show that Jaffé's theory of columnar recombination gives a good explanation of the pulse distribution in liquid argon. The average energy ω lost by an α particle during the process of producing an ion pair in argon gas has been determined by a number of workers (Bortner and Hurst 1953, Jesse and Sadanskis 1953, Genin 1956) using both polonium and plutonium as α sources. Agreement between the various investigations has been good and a value of 26·4 ev for ω can be considered reliable.

By measuring the distribution spectrum of pulses of electrons produced by the isotropic emission of α particles from a weak source of ^{239}Pu in liquid argon ω may be evaluated by a process analogous to that used in the gas phase. A determination of ω from single pulse data would seem preferable to a method requiring measurement of steady conduction currents (Ullmaier 1963), since in the latter case the source strength must be accurately known. Further complications would be introduced because the steady current is an average of the currents resulting from individual α particles which are emitted isotropically.

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2. Experimental details

The test cell and electrode arrangement used for the measurement of the distribution of electron pulses has been discussed in detail in a previous paper (Swan 1963). The argon was condensed at 90 °K and contained less than 0·005% impurity. No further purification was undertaken. A 0·02 μc deposit of ^{239}Pu on the surface of the cathode emitted α particles of 5·14 MeV into the liquid. The length of an α track in liquid argon is only about 5×10^{-3} cm, and thus there was no loss of charge due to the α particles passing laterally out of the active volume. The electrons escaping recombination within the ionized column cross the interelectrode spacing under the influence of the applied field and the resulting pulse is recorded after amplification on a 70 channel pulse-height analyser. The noise level at the input of the preamplifier was found to be 12 μv.

3. Experimental results and discussion

Figure 1 shows some typical distributions of pulse heights in liquid argon at a number of field strengths. It is clear that there is a considerable spread in pulse heights (or number of electrons arriving at the anode) from the various α particles. If there were no difference in the recombination in columns emitted at varying angles to the field and if the gap width were sufficiently large that the α-particle range was negligible in comparison the pulse spectrum should be a sharp peak with Gaussian noise from the amplifiers superimposed. Obviously this is not the case for the results shown in figure 1 and it is

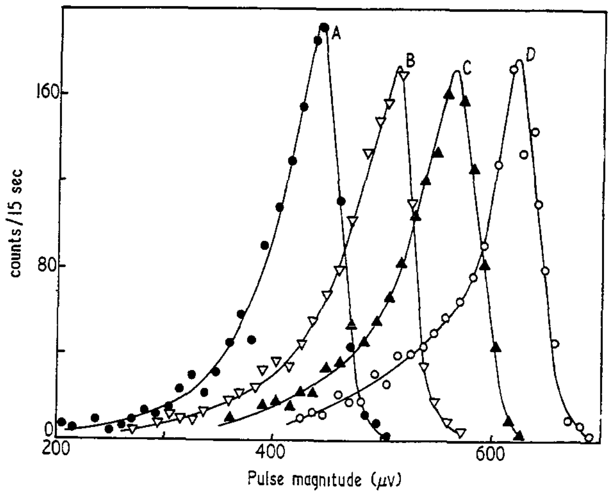

Figure 1. Pulse distributions in liquid argon. Electrode spacing, 0·117 cm. A, 34·2 kv cm^{-1}; B, 42·7 kv cm^{-1}; C, 51·3 kv cm^{-1}; D, 60·0 kv cm^{-1}.

reasonable to suppose that the distribution in pulse magnitudes is due either to recombination or to a geometric effect.

Assuming the emission of α particles from the cathode is isotropic, the fraction f of α particles emitted into an angle greater than the azimuthal angle θ will be given by

$$f = \cos \theta. \tag{1}$$

Now the geometric effect arises if the α-particle range is comparable with the gap width, since in that case the ions formed along the column do not have to traverse the whole gap. This has been discussed by Davidson and Larsh (1950) and it is sufficient to give here the theoretical variation of pulse height with angle θ for the case where l is the α-particle range and d is the gap width. This is

$$V(\theta) = \bar{V}\left(1-\frac{l}{d}\cos\theta\right)$$

where $\bar{V}$ is the maximum pulse height ($\theta = \pi/2$). It is assumed that all the ionization occurs at the end of the α track.

Introducing $\cos\theta$ from equation (1) gives

$$V(f) = \bar{V}\left(1-\frac{l}{d}f\right). \tag{2}$$

In order to derive the corresponding relationship for the case where recombination varies with angle of emission of the α particle it will be assumed that Jaffé's theory of columnar recombination is applicable. This assumption will be discussed later. According to the theory (Jaffé 1913, Loeb 1955) the number of ions set free from unit length of a column at an angle θ to the field after an infinite time is

$$n(E,\theta) = n_0\left\{1+\frac{\alpha n_0 S(E)}{4\sqrt{(2\pi)}b\mu E\sin\theta}\right\}^{-1} \tag{3}$$

where α is the recombination coefficient, b a linear dimension approximately the diameter of the ionized column, μ the mobility, and $S(E)$ a function of field which tends to unity for high field strengths. Now the voltage recorded due to $n(E,\theta)$ is $en(E,\theta)l/c\ (=V(E,\theta))$ where l is the column length, e the electronic charge and c the effective input capacitance of the measuring system. Similarly, the voltage induced by the ions in the absence of recombination would be $en_0l/c\ (=V_0)$. Inverting equation (3) and rearranging gives

$$\frac{1}{V(E,\theta)} = \frac{1}{V_0}+\frac{G}{E\sin\theta} \tag{4}$$

where

$$G = \frac{\alpha c S(E)}{4\sqrt{(2\pi)}b\mu e l}. \tag{5}$$

Introducing f from equation (1) into equation (4) gives

$$\frac{1}{V(E,f)} = \frac{1}{V_0}+\frac{G}{E(1-f^2)^{1/2}}. \tag{6}$$

If V_0 and G are known numerically the distribution may be obtained from equation (6).

The maximum pulse heights ($\bar{V}$) result from α particles emitted along the electrode surface ($\theta = \pi/2$) and thus the relationship between these maximum pulse heights and the field is given from equation (4) as

$$\frac{1}{\bar{V}(E)} = \frac{1}{V_0}+\frac{G}{E}. \tag{7}$$

A plot of $1/\bar{V}(E)$ against $1/E$ should therefore be linear at high fields since $S(E)$ in G

(equation (5)) tends to unity. The slope of this characteristic is G and the extrapolated intercept V_0^{-1}. Inserting these values into equation (6) then enables the distribution of pulses at any field strength to be predicted.

Figure 2 shows the results obtained for two different electrode spacings over a field range from 10 kv cm^{-1} to 60 kv cm^{-1} and plotted according to equation (7). At the

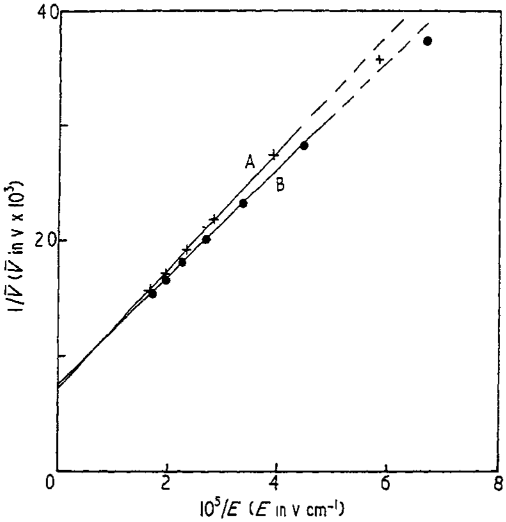

Figure 2. Maximum pulse heights in liquid argon plotted according to equation (7). A, d = 0·117 cm, c = 23·0 pF; B, d = 0·0666 cm, c = 23·9 pF.

higher field strengths the linearity is good enabling both G and V_0 to be calculated. From these results G is found to be $5{\cdot}34 \times 10^7$ cm^{-1} and V_0 is $1{\cdot}39 \times 10^{-3}$ v for a gap length of 0·117 cm. The distributions of pulse heights at three field strengths have been computed from equation (6) using these values and are shown in figure 3. The

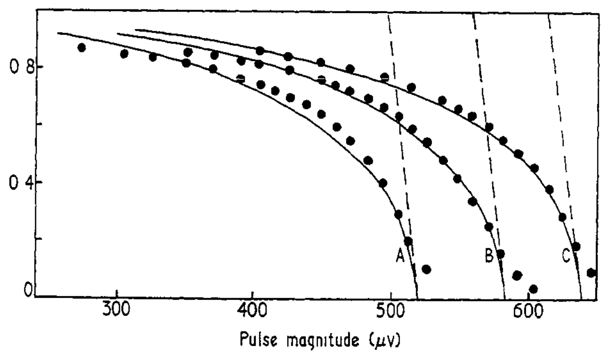

Figure 3. Pulse distributions in liquid argon. Full curve, theoretical distribution from equation (6); broken curve, theoretical distribution from equation (2); •, experimental points; A, 42·7 kv cm^{-1}; B, 51·3 kv cm^{-1}; C, 60·0 kv cm^{-1}.

experimental points are taken from figure 1. Also shown in figure 3 are the distributions resulting from a geometrical effect for the case in which recombination is independent of θ. The α-particle range l is taken as 5×10^{-3} cm. The agreement between the experimental results and equation (6) indicates that recombination is responsible for the distribution of pulse magnitudes, and also that Jaffé's theory is applicable to the case of α-particle ionization of liquid argon. If the magnitude of the electrode spacing is such that the geometric effect is significant this correction should be subtracted from the theoretical distribution derived on a basis of recombination loss.

From the extrapolated values of V_0 the average energy ω required to form an ion pair may be evaluated from the relationship $\omega = \epsilon e/cV_0$, where ϵ is the energy of an α particle. From the two characteristics in figure 2, ω is found to be 25·6 ev and 26·4 ev. Because of the inherent uncertainty involved in the extrapolation, the difference between the average value of 26·0 ev in the liquid and that of 26·4 ev for argon gas is not significant, and it would seem that as far as energy loss is concerned the collisions involved in the retardation of an α particle in liquid and gaseous argon are the same.

The gradient G contains many factors the numerical values of which are not known with any certainty. In particular, the recombination coefficient for electrons and argon ions has not been measured in the liquid state and the diameter of the ionized column can only be estimated. However, since the behaviour of electrons in liquid argon is in many respects analogous to that of electrons in gaseous argon it is reasonable to suppose that the value of α ($6{\cdot}7\times10^{-7}$ cm^3 sec^{-1}), measured for the recombination of electrons with $Ar_2{}^+$ ions in the gaseous phase (Oskam and Mittelstadt 1963), is also applicable to the recombination in the liquid phase. For the examples given in figure 2 the mean gradient G is 5×10^7 cm^{-1}, and with a capacitance c of 23 pF equation (5) gives $b\mu \simeq 4\times10^{-5}$ cm^3 v^{-1} sec^{-1}. Now the mobility of positive ions in liquid argon ($\simeq10^{-4}$ cm^2 v^{-1} sec^{-1}) (Davis, Rice and Meyer 1962) is very much less than that of electrons ($\simeq10$ cm^2 v^{-1} sec^{-1}, (Swan 1964)), but since the rate of separation of charges and the time during which recombination within the column can occur is determined by the larger of these mobilities a measure of the column diameter can be obtained by substituting for μ a value of 10 cm^2 v^{-1} sec^{-1}. This gives $b \simeq 4\times10^{-6}$ cm, in good agreement with previous estimates (Kramers 1952).

There has been much speculation as to the applicability of Jaffé's columnar recombination theory to liquids, particularly when the ionizing radiations are α particles (Stacey 1958, Kramers 1952). For liquids in which the electrons attach rapidly to the liquid molecules to form negative ions the criticisms put forward would seem justified, and Kramers' (1952) modified theory, which assumes that recombination loss is dominant and that diffusion contributes only a small correction, does give a better explanation for certain liquids at low fields (Gerritsen 1947). However, in liquid argon, where the electrons remain free, the contribution of diffusion would be expected to predominate in the separation of the ion species and in particular the greater diffusion coefficient of the electrons would be the determining factor in the number of carriers escaping from the column. Thus Jaffé's theory, which assumes diffusion to be the significant parameter, might be expected to apply to liquid argon and the results presented in this paper support this view.

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