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The Ratio of Diffusion Constant to Mobility for Electrons in Liquid Argon --- An Evidence of Non-existence of the Ramsauer Minimum in Liquid Argon ---

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The ratio of diffusion constant(D) to mobility(μ) for electrons has been measured in liquid argon by using modified Townsend's apparatus. The measured ratio of D/μ monotonously increases from 0.1 to 0.4 V with increase of the electric field(E) from $\sim$2 to $\sim$10 kV/cm. In this region of E, D/μ is smaller than that obtained in gaseous argon, compared at the same value of E/N, where N is the atomic density of argon. Momentum transfer cross section is estimated to be almost constant (about $3 \times 10^{-17} cm^2$) for the electrons with the energy less than $\sim$0.3 eV, and for the electrons with larger energy, shows the tendency to increase. This shows that the Ramsauer minimum no longer exists in liquid argon.

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I INTRODUCTION

Electron drift velocities in liquefied rare gases have been measured for over twenty years. Ten years ago, Miller et al.[1] extensively measured the drift velocities of electrons in condensed states of rare gases, with attension to eliminating the effect of impurities. Recently, our group[2] and Yoshino et al.[3] studied the effects of molecular impurities to the electron drift velocities in the rare gas liquids. In their experiments, a considerable increase in electron drift velocity was found. Such an effect is well known in the gas phase[4,5], and is explained by the reduction of mean electron energy due to the inelastic collision of electrons with molecular impurities, and by the decrease of momentum transfer cross section (Ramsauer-Townsend effect). For liquid argon, however, the magnitude of increase in electron drift velocity is less significant than for gaseous argon. This fact might suggests the lack of Ramsauer effect in the liquid[2].

In 1967, Cohen and Lekner[6] gave a solution of the Boltzmann equation for the electrons in gases, liquids, and solids. And Lekner[7] applied the solution to the electronic motion in liquid argon and gave basic transport parameters for electrons, such as drift velocity (w), mean agitation energy of electrons ($\langle \varepsilon \rangle$), momentum transfer mean free path (λ), etc. as a function of electric field strength (E). The principal result is that the Ramsauer minimum no longer exists in liquid argon.

In liquid argon, however, no experimental evidence has been

presented directly showing the lack of Ramsauer minimum. The situation is that, the electronic motion under the electric field is, in principle, determined by the agitation energy, ε, of electrons and by the cross section, $\sigma(\varepsilon)$, as a function of ε, but the information obtained from the experiments so far is only on w and not on ε except the trial of Derenzo[8]. Concerning the information on ε, the ratio of diffusion coefficient (D) to mobility coefficient (μ) plays an important role. As seen from the so-called Einstein's relation,

$$eD/\mu = kT, \qquad (1)$$

the ratio eD/μ is a good measure of $\langle\varepsilon\rangle$[9], where e is the electronic charge, k is Boltzmann's constant and T is the absolute temperature.

In this paper, we present the data on the ratio, eD/μ, as a function of E for electrons in liquid argon, obtained by the modified Townsend's method using a parallel plate pulse ionization chamber. The results were compared with those calculated by Lekner and with those of Derenzo's trial. Furthermore, the momentum transfer cross section is discussed in comparison with that in gaseous argon.[10-12]

II. PRINCIPLE OF MEASUREMENT ON eD/μ

In the measurement, parallel plate pulse ionization chamber designed as a modified Townsend's apparatus was used. Here, we consider an ideal isolated travelling group of n_o electrons from the cathode to the collector under the influence of uniform and constant electric field (E) in the chamber. The group having the point-like distribution at the position $(x,y,z) = (0,0,0)$ at the time $t = 0$, is described in the following form[9] at the time of t,

$$n = \frac{n_o}{(4\pi Dt)\ (4\pi D_L t)^{1/2}} \left[\exp\left(-\frac{\rho^2}{4Dt}\right)\right] \exp\left[-\frac{(z-wt)^2}{4D_L t}\right] \quad ,(2)$$

where z-axis is chosen to be the direction of E, w is the drift velocity of the group, D is the coefficient of diffusion concerned with lateral diffusion to E, D_L is the longitudinal coefficient of diffusion, and $\rho^2 = x^2+y^2$. Here, the average of ρ^2 is easily obtained to be 4Dt, then 4Dt is a measure of lateral dimension of the cloud.

In the configuration shown in Fig.1, the number of electrons, n_{k-1}, n_k, n_{k+1}, collected by each separated collector, c_{k-1}, c_k, c_{k+1}, are independent on D_L, under the condition that the distance between the collector and the cathode (d) is sufficiently larger than the longitudinal dimention of the cloud, $\sqrt{4D_L\tau}$ (that is, $\sqrt{D_L\tau} \ll d$) where $\tau = d/w$. Then, we can also assume for convenience in calculation that $D_L = D$. Thus, the distribution in the group in the arrival at the collector is written as

follows,

$$n(r) = n_o \exp\left(-r^2/R^2\right)/(\pi^{1/2}\cdot R)^3 \qquad (3)$$

where $r^2 = x^2+y^2+(z-w\tau)^2$, and

$$R^2 = 4D\tau. \qquad (4)$$

Now, we can calculate the number of electrons, n_k, arrived at the collector, c_k, occupying the region from X-s/2 to X+s/2 along x-axis, (having the center at X with the width of s along x-axis), sufficiently long along y-axis, and d along z axis. The number is written as follows,

$$n_k = 2\pi\left[\int_{X-s/2}^{X+s/2} r\cdot(r-X+s/2)\cdot n(r)\,dr + \int_{X+s/2}^{\infty} r\cdot s\cdot n(r)\,dr\right]. \qquad (5)$$

The variation curves of the ratio, n_k/n_o, obtained from eq. (5) are shown in Fig.2 for some typical values of R, as a function of the ratio X/s in the case of s = 60μm, which corresponds to the condition of present experiment as will be described in Sec.III. The calculated curves of the ratio are fitted to experimental data by adjusting R and X as parameters, and thus the value of R is determined.

Finally, we can obtain the ratio, eD/μ, from the following relation,

$$eD/\mu = e(R^2/4\tau)\cdot(E/w) = eR^2V/(4d)^2, \qquad (6)$$

where V is the applied voltage between the cathode and the collector.

III. EXPERIMENTAL APPARATUS AND PROCEDURE

In this experiment, a pulse ionization chamber filled with liquid argon having the split collectors and a cathode, on which a narrow line-like source of alpha-particles was deposited, was used, and the determination of R was made by measuring the charge collected by each individual collector.

A. Ionization chamber

The cathode is a flat-surfaced stainless-steel plate with the effective area of 25 x 25 mm^2. The fabrication of the source of alpha-particles is as follows; first, ^{210}Po was deposited through a photo-etched rectangular hole (15 μm x 1 mm) at the center of a plastic mask coated on the cathode, and after that, the mask was solved off by chemical etching. Thus obtained source of ^{210}Po was confirmed to be less than 20 μm in width, by auto-radiography of X-ray film.

As shown in Fig.3, the collector, which has the effective area of 24 x 24 mm^2, consists of sixteen strip collectors ~~electrodes~~ of 20 μm in width with the spacings of 10 μm and two outer electrodes of 12 mm in width. These collectors ~~electrodes~~ are gold films deposited on ceramic plate through a plastic mask formed by

photo-etching. All 18 portions of the collector are insulated from each other inside the chamber. In the present experiment, these ~~electrodes~~ collectors were externally connected and were divided into 8 parts named a, b, c, d, e, f, g, h, as shown in Fig.3. Thus, the ~~electrodes~~ collectors in the measuring parts, b, c, d, e, f, g, effectively have the width of 60 μm.

During the experiment, the distance between the cathode (K) and the collector (C) was varied from 1.7 mm to 4.7 mm. Under these conditions, the electric field in the region of electron drift was sufficiently uniform without shaping electrode of field (guard-ring).

Commercial tank argon of 99.999% purity was further purified by the purifier with barium-titanium getter used in our previous experiments[2,13]. The ionization chamber was inserted into a vacuum tight vessel made of stainless-steel and cupper, and then was baked out at 100°C and evacuated to a pressure less than 1.0×10^{-7} Torr with the gas filling system. The purified argon was condensed into the vess by cooling down with liquid oxigen refrigerant.

B. Electric circuit

Signals induced on the ~~above electrodes~~ strip collectors were amplified by a charge-sensitive preamplifier and fed to a mainamplifier with the differential and integral time constants of 2 μsec, and analyzed by a multichannel pulse height analyzer. To reduce the

background noise, the pulses from the collectors other than the collector (c_i) nearest to the center line of the alpha-source were gated by using the largest pulses obtained from the collector c_i, as shown in Fig.4. Pulses corresponding to the total electrons n_o of the group were measured by the electrical connection of all ~~electrodes~~ collectors. Thus, the ratios, n_{i-1}/n_o, n_i/n_o, n_{i+1}/n_o, were obtained.

IV. RESULTS AND DISCUSSION

An inserted figure in Fig.5 shows a typical pulse height spectrum of ^{210}Po source, obtained with the external connection of all collectors. Curves of total collected charge against the electric field strength are shown in Fig.5 for different distances (d) between the cathode and the collector. As seen from the figure, the ~~fact that~~ pulse height does not depend on d which result in reduction of This shows that electronegative impurities, ~~for reducing the~~ pulse heigh in liquid argon are sufficiently removed by our purification method

Figure 6 shows the relation between d and the square of R obtained by the fitting method described in Sec.II. In Sec.II, we assumed that ~~it~~ the group of electrons had a point-like distribution at the starting point of the drift. Here, let us consider a group of electrons which have the Gaussian distribution as written in the following form at t = 0,

$$n(r) = n_o \exp(-r^2/R_o^2)/(\pi^{3/2} R_o^3).$$

Then, eq.(4) is modified as follows,

$$R^2 = 4D\tau + R_o^2. \qquad (7)$$

From eq.(7) and Fig.6, we can estimate the value of R_o to be ~10 μm, by extrapolating τ to zero. And then, the relation (6) is rewritten as,

$$eD/\mu = e(R^2-R_o^2)V/(4d^2). \qquad (8)$$

Actually, R_o is determined by the range of alpha-particles isotropically emitted from ^{210}Po and by the spread of ^{210}Po on the cathode.

The effect of positive ions on the drifted electrons can be neglected, because d is sufficiently larger than the range of alpha-track in liquid argon. The space charge effect of electrons in the diffusion measurement is more complicated. Here, we shall estimate only the mutual repulsion of electrons due to the effect of space charge in the absence of electronic diffusion, by applying the one dimensional treatment of McDaniel[4] to three dimensional one. Let us consider a group of n_o ~~the number of~~ electrons n_e which distribute uniformly on the surface of a sphere with a radius of r_o at the time t=0. By Gauss's theorem the field intensity outside the surface at the point which is at a distance of r from the center of the sphere is then $E=n_o e/(4\pi\varepsilon_A r^2)$, where ε_A is the dielectric constant of liquid argon. Electrons at the surface move under the influence of

this field with a drift velocity of $w = \mu E$, where μ is the mobility of electron. Then the spread of the sphere is written as follows, $dr/dt = \mu n_o e/(4\pi\varepsilon_A + r^2)$, and the solution is easily obtained as $r = [\, 3\mu n_o e t/(4\pi\varepsilon_A) + r_o^3 \,]^{1/3}$. In the result, the size of the sphere increases after the drift of d, up to the radius, R_s, written as

$$R_s^2 = [\, 3\mu n_o e \tau/(4\pi\varepsilon_A) + r_o^3 \,]^{2/3}, \qquad (9)$$

where $\tau = d/w$.

In principle, the present results on R should contain the contribution from the space charge effect as shown in eq.(9). However, the experimental results show the linear relation between R^2 and d as seen from Fig.6, and the subtraction of the effect of the space charge, which is non-linear for d, from experimental results leads to the unreasonable situation that $R^2 - R_s^2$ is also non-linear for d. ~~Here,~~ So, we assume that the effect of the electron space charge is negligibly small.

In Fig.7, the ratios of eD/μ thus obtained from eq.(8) for E are plotted, and the ratios of eD/μ obtained by Derenzo[8] in liquid argon are also plotted. It is expected that the effect of space charge more or less increases the apparent ratios of eD/μ in our experiments and decreases those of Derenzo's experiments, because in order to produce electrons he used high-energy charged particles penetrating the detector parallely to the electric field and the produced electrons and ions were attracting each other throughout the drift of electrons. However, the agreement between ours and Derenzo's

is fairly good within the experimental error. This fact also suggests that the effect of space-charge is small compared with the experimental error.

Warren and Parker's result for gaseous argon[10] is compared in Fig.7 with those for the liquid, where $N = 2.1 \times 10^{22}/cm^3$ is used as the atomic density in liquid argon. The values of $\langle\varepsilon\rangle$ theoretically obtained by Lekner are also plotted in the figure by ~~the~~ use of the approximate relation, $eD/\mu = 2\langle\varepsilon\rangle/3$. This relation can rigorously be applied only to the electrons with Maxwellian distribution, but, in practice, will be also valid for other probable distributions for the rough estimation[9]. From the figure, we can conclude that the electronic temperature in liquid argon is lower than that in gaseous argon ~~in~~ over the ~~extent~~ region of E/N used in this experiment and that the experimental result is in agreement with that of Lekner rather than that in gaseous argon. The difference in eD/μ for the different d is negligibly small, which corresponds to the fact that eq.(7) is well established as shown in Fig.6.

From the results shown in Fig.7 and the electron drift velocities obtained in liquid argon[1], we can estimate the momentum transfer cross section of electrons, σ, by the use of the formula $\sigma = eE/[Nw(2m\langle\varepsilon\rangle)^{1/2}]$, derived from the relation $w = Ee\lambda/(m\langle v\rangle)$, which is used by McDaniel[4], where m is the mass of electrons, $\langle v\rangle$ is the average velocity of electrons, λ is the mean free path, and $\langle\varepsilon\rangle = m\langle v\rangle^2/2$. Thus obtained values of σ against $\langle\varepsilon\rangle$ are plotted in Fig.8. The values of σ for smaller

$\langle\varepsilon\rangle$ (nearly 0.01 eV) are obtained by using the values of the electron mobility[1,3,14] at low-field, where the electronic temperature is regarded to be equal to that of liquid argon. The solid curve in Fig.8 is obtained by using the experimental data of w[15] and eD/μ[10] for gaseous argon obtained with the same manner as that for the liquid as described above. The momentum transfer cross sections obtained from the mean free path calculated by Lekner are also plotted in the figure. They are in agreement with the present experimental ones. From thus obtained curve, the outline of the variations of σ against $\langle\varepsilon\rangle$ in both phases of argon ~~can be seen well~~ is known, although the above derivation of σ is not so rigorous especially when σ varies with $\langle\varepsilon\rangle$. For gaseous argon, the dip of σ in $\langle\varepsilon\rangle$ of about 0.4 eV corresponds to Ramsauer-Townsend effect. On the other hand, for liquid argon, σ is almost constant for $\langle\varepsilon\rangle$ smaller than ~0.4 eV. This shows that Ramsauer minimum no longer exists in liquid argon as pointed out by Lekner.

V. CONCLUSION

From the present experiment made by the modified Townsend method, we can deduce the conclusions described below.

In liquid argon, the value of eD/μ for electrons, which is a good measure of mean electron energy, monotonously increases from ~0.1 eV to ~0.4 eV with increase in the electric field from ~2 kV/cm up to ~10 kV/cm. In this region of electric field, eD/μ is smaller than that in gaseous argon, that is, the

electron temperature in the liquid is lower than that in the gas at the same E/N.

Momentum transfer cross section is almost constant (3×10^{-17} cm^2) in the electron energy region from ~0.01 eV up to ~0.4 eV in liquid argon. On the other hand, the cross section in gaseous argon have the distinct dip at the electron energy near 0.3 eV as the result of Ramsauer-Townsend effect. This shows that the Ramsauer-minimum no longer exists in liquid argon.

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FIGURE CAPTIONS

Fig.1 Schematic drawing which shows the diffusion process of drift electrons in a gap between parallel plate electrodes. The names c_{k-1}, c_k, c_{k+1},..., are given to the split collectors, respectively, and n_{k-1}, n_k, n_{k+1},..., express the number of electrons collected by c_{k-1}, c_k, c_{k+1},..., respectively. For example, n_k is equal to the number of electrons contained in the volume between the plains, $x = X-s/2$ and $x = X+s/2$.

Fig.2 Typical distribution of the relative number of electrons collected by each collector of 60 μm in width agaist a distance from the center of electron group.

Fig.3 Photographs of collector electrode.

(a) Whole electrode

(b) Magnified view of the area shown with white circle in (a), and electrical connections.

Fig.4 Schematic diagram of electronic circuits. This figure shows that the number of electrons collected by the collector c_{i+1} is measured by gating with the pulses from c_i.

Fig.5 The variation of total collected charge due to ^{210}Po versus electric field E, for different distances (d) between the cathode and the collector electrodes. Inserted figure shows a typical pulse height spectrum, when d = 1.7 mm and E = 5.9 kV/cm.

Fig.6 Square of the measured mean width of electron distribution R^2 against the drift length d for several values of

electric field strength E.

Fig.7 Field dependence of the ratio, eD/μ in liquid argon. The points represented by ○, ●, ×, ✱, are the present results. The points □, △ are the results according to Derenzo[8], and Lekner[7], respectively. Solid curve shows the results for gaseous argon[10].

Fig.8 Variations of approximate momentum transfer cross section δ as a function of mean electron energy $\langle\varepsilon\rangle$. The points represented by ○, ●, ×, ✱, □, △, are corresponding to the results shown by these marks in Fig.7. The points represented by ■, ▽, ▲, correspond to the results in the Ref. 1), 3), 14), respectively.

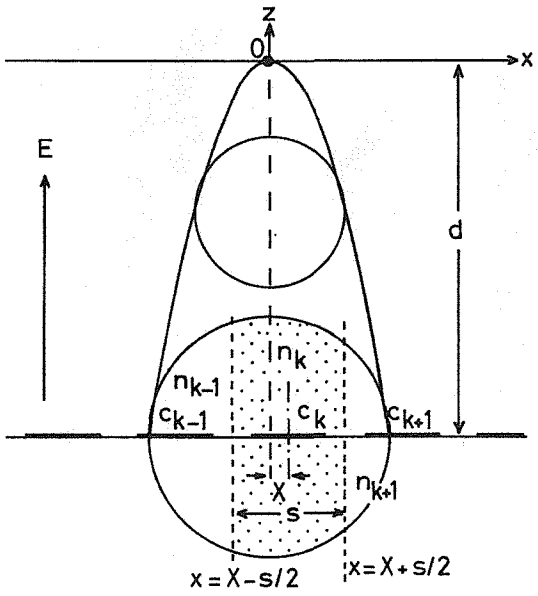

z
0
x
E
d
n_k
n_{k-1}
c_{k-1}
c_k
c_{k+1}
X
s
n_{k+1}
x = X - s/2
x = X + s/2

Fig. 1

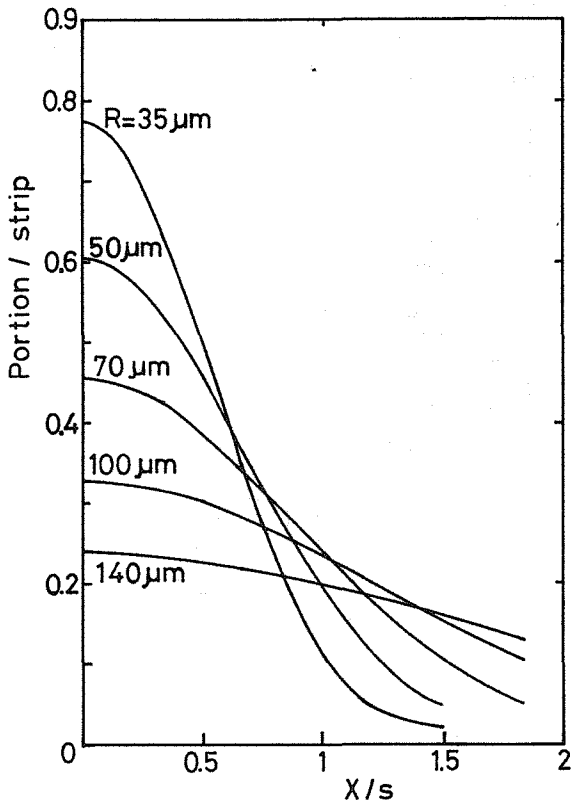
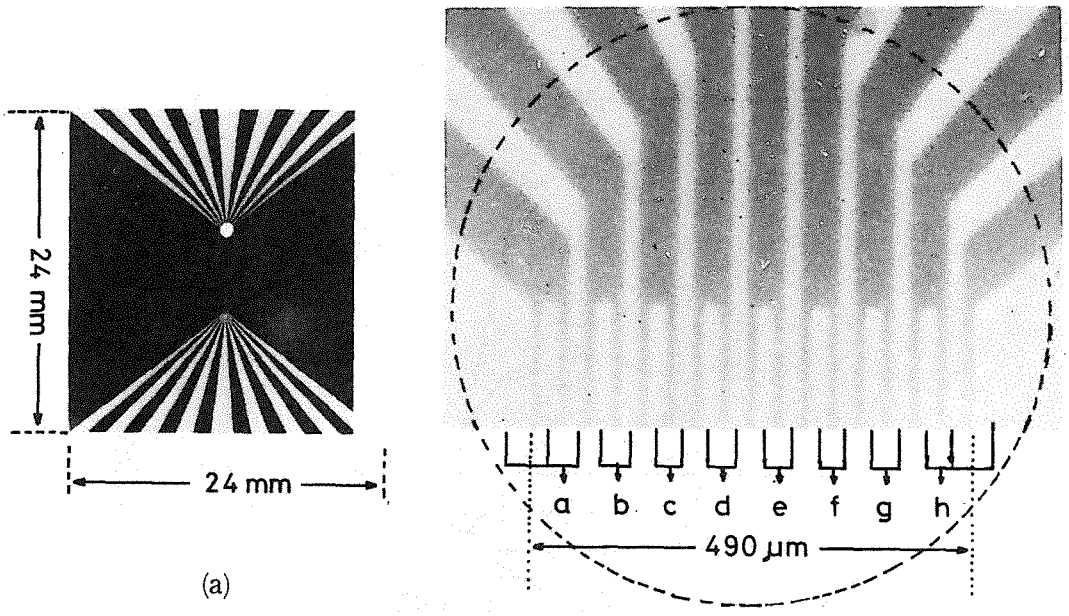

0.9
0.8
0.6
0.4
0.2
0
R=35 μm
50 μm
70 μm
100 μm
140 μm
Portion / strip
0.5
1
1.5
2
X / s

Fig. 2


24 mm
24 mm
a b c d e f g h
490 μm

(a) (b)

Fig. 3

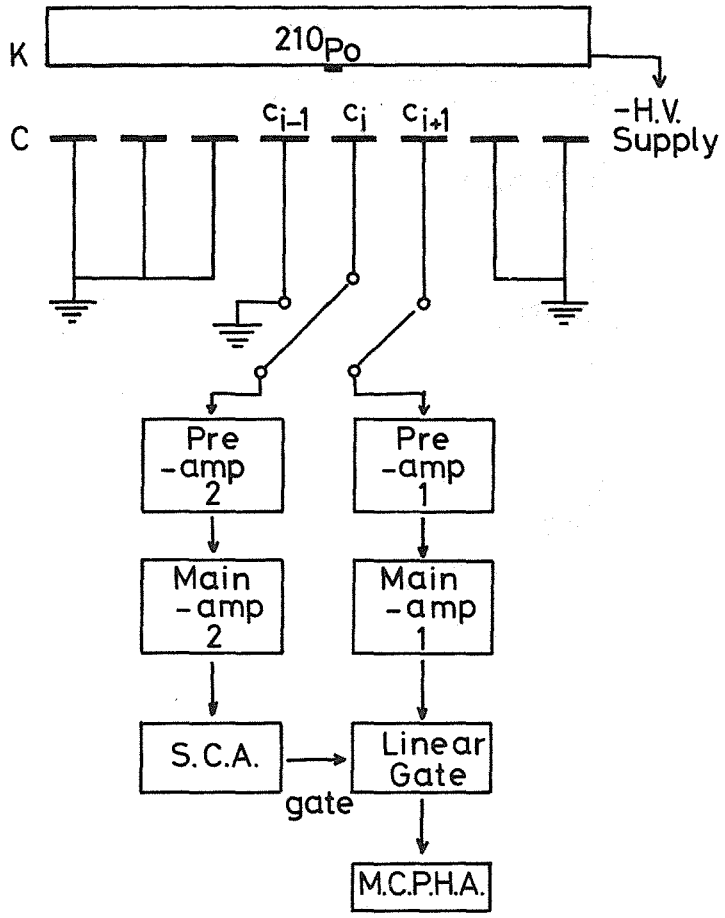

K
210Po
C
ci-1 ci ci+1
-H.V. Supply
Pre -amp 2
Pre -amp 1
Main -amp 2
Main -amp 1
S.C.A.
Linear Gate
gate
M.C.P.H.A.

Fig. 4

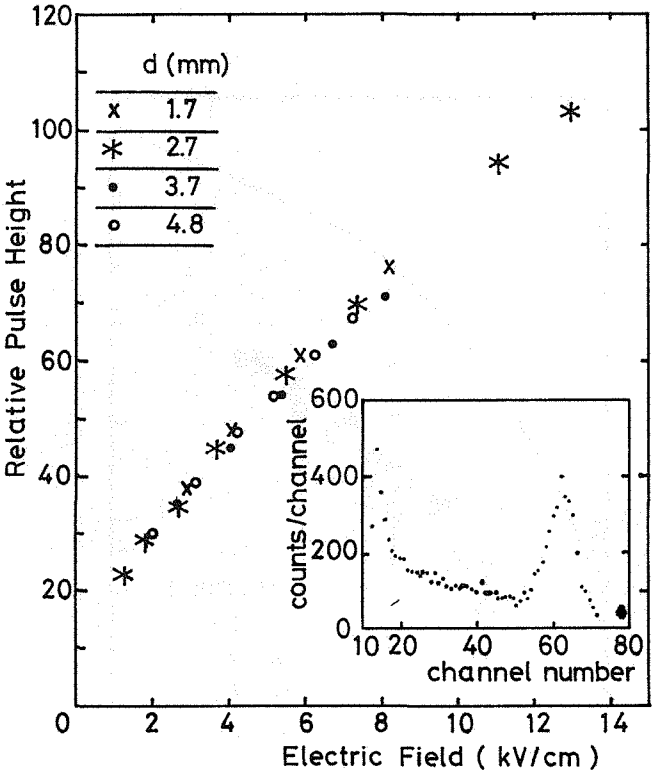

d (mm)
x 1.7
* 2.7
• 3.7
o 4.8
Relative Pulse Height
Electric Field (kV/cm)
counts/channel
channel number

Fig. 5

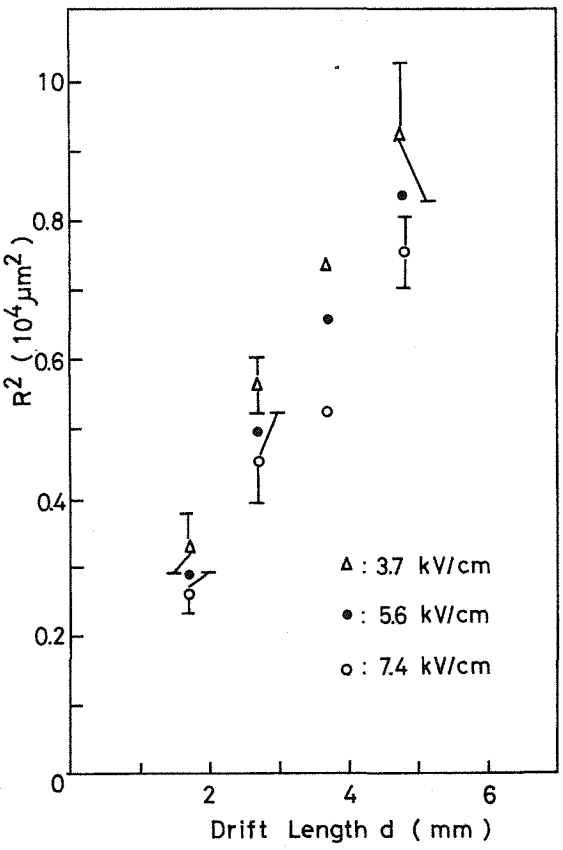

R² (10⁴μm²)
Drift Length d (mm)
Δ : 3.7 kV/cm
• : 5.6 kV/cm
o : 7.4 kV/cm

Fig. 6

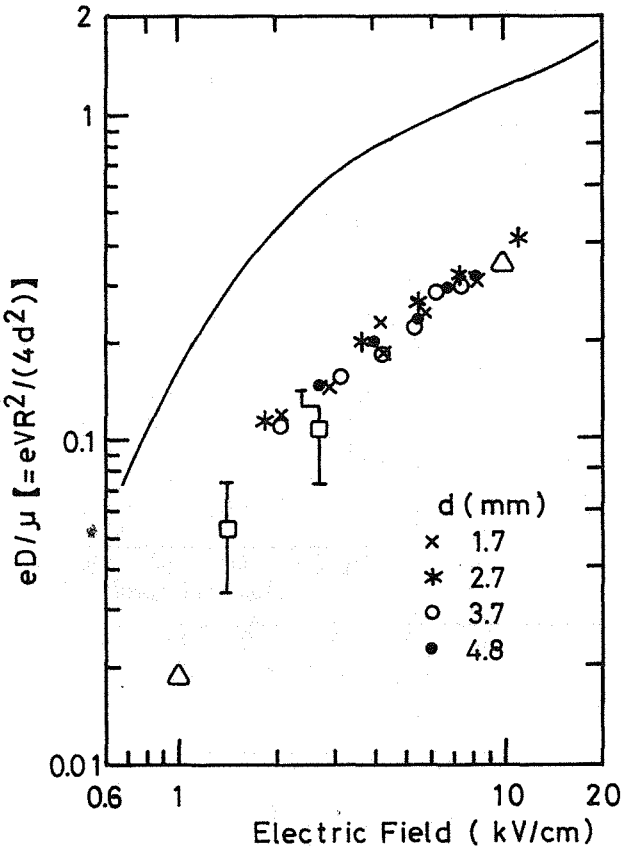

2
1
0.1
0.01
0.6
1
10
20
eD/μ [=eVR²/(4d²)]
Electric Field (kV/cm)
d (mm)
× 1.7
* 2.7
○ 3.7
● 4.8

Fig. 7

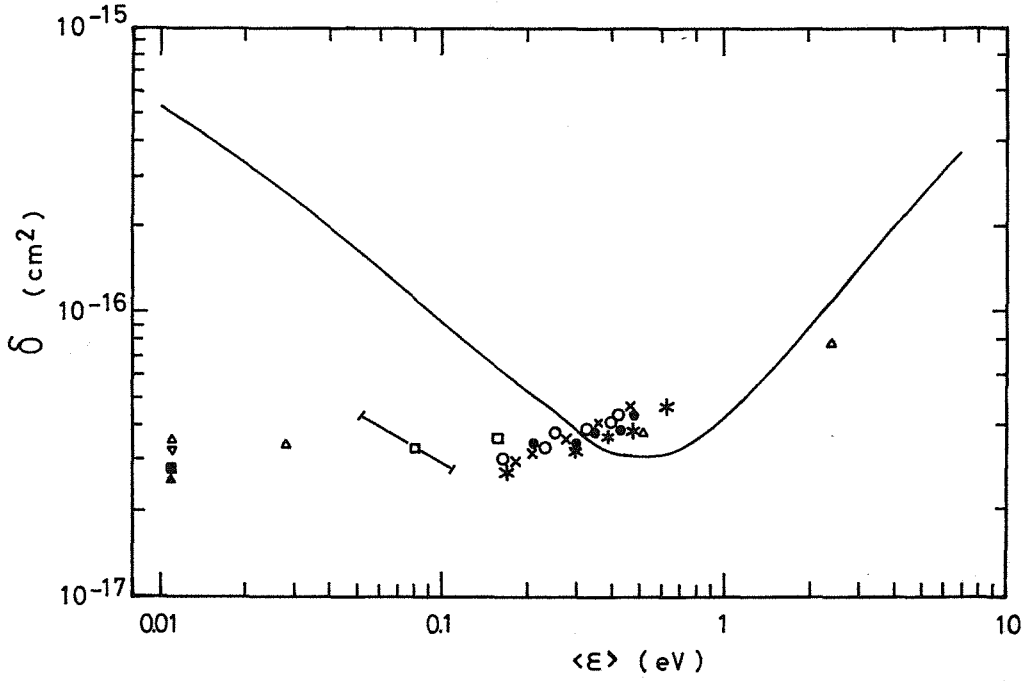

10⁻¹⁵
10⁻¹⁶
10⁻¹⁷
δ (cm²)
0.01
0.1
1
10
⟨ε⟩ (eV)

Fig. 8