Scintillation and ionization yield for α particles and fission fragments in liquid argon

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(Received 30 July 1986)

The scintillation and ionization yields in liquid argon by α particles and fission fragments have been measured as a function of the applied electric field (≤ 38 kV/cm). The quenching due to high excitation density does not depend on the field for $\gtrsim 4$ kV/cm for α particles. The scintillation due to α particles shows a maximum at about 1.5 kV/cm. This increase ($2.5\pm0.7\%$) of the scintillation at low fields has not been observed heretofore and is ascribed to the presence of a quenching process at zero field.

The behavior of the scintillation and ionization yields in liquid argon under an electric field has been studied before for electrons[1] and relativistic heavy ions,[2] and the results show that the total energy deposited T is expressed by a linear combination of the scintillation S and ionization Q yields, i.e., $T=Q+aS$, where a is a constant.[2] The discovery of this linear relation has initiated the development of the liquid-phase photoionization detector[3,4] which can be a powerful tool for studies of high-energy heavy ions. We have extended the study for α-particle and fission-fragment excitation, which involves a quenching process due to high excitation density.

The apparatus used in the experiment is an ionization chamber with a Pyrex glass window for observation of scintillation light, and is in essence the same as that reported before.[4] A thin source of α particles (^{210}Po or ^{252}Cf), or fission fragments (^{252}Cf), was deposited on a stainless-steel cathode 3.8 cm in diameter and a grid-type collector, which transmits 80 or 90% of the light, is located 2.1 mm from the cathode. The scintillation in liquid argon is in the vacuum ultraviolet (vuv) region. This light is converted to visible light by a sodium-salicylate wavelength shifter and detected by a photomultiplier (Hamamastu R329). The charge signals from the collector were fed into a charge-sensitive preamplifier that in turn was connected to a main amplifier (Canberra 2010) whose shaping time constant was set at 4 μsec. The charge signals for fission fragments were gated by the scintillation signal since the α peak and the fission peaks are superposed.

The ultimate vacuum obtained for the ionization chamber before filling was about 4×10^{-7} Torr, and its outgassing rate was less than 5×10^{-5} Torr/h. Argon containing impurities less than 10 ppm was further purified using a barium-titanium getter before being condensed into liquid argon.

The scintillation and ionization yields obtained for α-particle and fission-fragment excitation are plotted as a function of applied electric field in kV/cm in Fig. 1. The ionization yields increase slowly with increasing electric field E. As much as 26% of the total charge produced by a 6.12-MeV α particle was collected at $E=38$ kV/cm, assuming that the W values are the same for electrons (23.6 eV) (Ref. 5) and α particles. The charge collected for a 5.305-MeV α particle was slightly lower as shown in Fig. 1. The decrease in the scintillation yields with increasing electric field was almost the same for both of these α particles and was 23% below the value observed at zero electric field for a 6.12-MeV α particle at $E=38$ kV/cm. An increase over the general trend in the scintillation yield at low electric field was observed for α particles. Such an increase has not been observed for electrons and relativistic heavy ions.[2] The maximum increase observed is $2.5\pm0.7\%$ at about $E=1.5$ kV/cm while the charge collected with this field is about 3%.

The charge collection and the resolution were rather poor for fission fragments, and peaks due to the heavy and light fragments were not resolved. Therefore, we

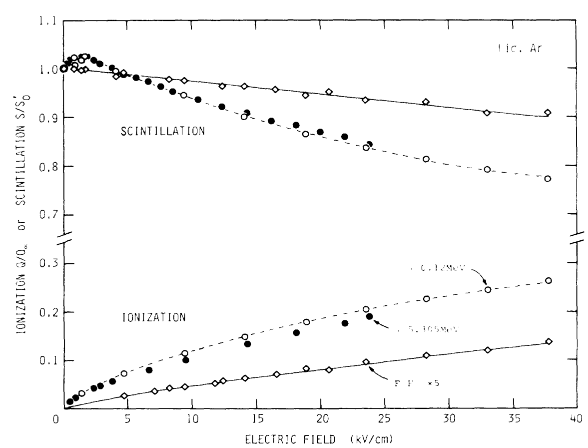

FIG. 1. Variation of the scintillation S and ionization Q yields for α particles (●, 5.305 MeV; ○, 6.12 MeV) and fission fragments (FF) (◇) in liquid argon as a function of applied electric field. S_0' is the scintillation yield obtained at zero electric field, and Q_∞ is the charge produced by an ionizing particle.

took the pulse height for the center of gravity; the error in estimating energy deposited due to this procedure is about $\pm 15\%$. The charge collected can be expressed roughly as $CE^{0.80}$ where C is 0.15% or 9.4×10^{-16} C and E is in kV/cm in the region of $4 \lesssim E \lesssim 38$ kV/cm. The charge collected was less than 3% at $E = 38$ kV/cm. The charge may include some contributions from prompt γ rays. However, these contributions should be small since the increase in charge collected is a slow function of the electric field. The charge collection is much more efficient for electrons and γ rays.[5] The scintillation yield plotted in Fig. 1 for fission fragments is the average value for light and heavy fragments. The decrease in scintillation yield observed at $E = 38$ kV/cm was 9% of the value at zero electric field. The increase in scintillation observed for α particles at low electric fields was not detected for fission fragments within the experimental error ($\pm 1\%$).

In the presence of quenching, as we assume here, both the exciton and recombination light are quenched in the same way. Then, the number of ion pairs and excitons available for scintillation and charge collection is $q(N_i + N_{ex})$, where q is the quenching factor, and q does not depend on the electric field E. We assume also the fraction of electrons escaping from recombination χ is zero. Then the number of ion pairs N_i and excitons N_{ex} produced by a charged particle is expressed by the equations

$$N_i + N_{ex} = (q\phi_{vuv})^{-1} S'_0 \quad \text{for } E = 0 , \tag{1}$$

$$N_i + N_{ex} = q^{-1} Q + (q\phi_{vuv})^{-1} S \quad \text{for } E \neq 0 , \tag{2}$$

where Q is the charge yield measured in electrons, and S and S'_0 are the numbers of photons produced in the presence and absence of the electric field, respectively. The number of photons expected in the absence of quenching is $S_0 = S'_0 / q$. ϕ_{vuv} is the quantum efficiency for vuv emission. From Eqs. (1) and (2), we have

$$\frac{S}{S_0} = \frac{q(1 + N_{ex}/N_i) - Q/Q_\infty}{1 + N_{ex}/N_i} , \tag{3}$$

where Q_∞ $(= N_i)$ is the total charge produced by an ionizing particle. Setting $q = 1$ and adding $-\chi$ into the denominator, we have a linear relation between S and Q, as obtained previously in the absence of the quenching, i.e.,[6]

$$\frac{S}{S_0} = \frac{1 + N_{ex}/N_i - Q/Q_\infty}{1 + N_{ex}/N_i - \chi} . \tag{4}$$

Data taken for electrons and relativistic Ne and Fe ions lay on a straight line given by the equation with $N_{ex}/N_i = 0.21$ and with $\chi = 0.35$ and 0, respectively.[6]

The result obtained for α particles is plotted in Fig. 2. Quenching factors q at zero electric field were found to be 0.71 ± 0.04 and 0.73 ± 0.04 for 5.305- and 6.12-MeV α particles, respectively, and 0.17 ± 0.02 for fission fragments in liquid argon in the absence of the electric field.[2,7] The main contributions to the uncertainty are systematic errors in comparing scintillation yields for α particles and relativistic heavy ions.[2] The solid line shows Eq. (3) with $N_{ex}/N_i = 0.21$ and $q = 1$, i.e., Eq. (4) with $\chi = 0$. If the

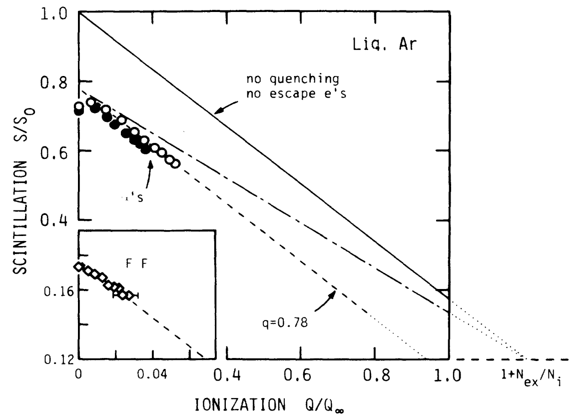

FIG. 2. Relation between scintillation S/S_0 and ionization Q/Q_∞ yields obtained for α particles (●, 5.305 MeV; ○, 6.12 MeV) and FF (◇) in liquid argon. S_0 is the scintillation yield expected in the absence of quenching.

quenching does not depend on the electric field, Eq. (3) gives a straight line parallel to the solid line. Dashed lines show Eq. (3) with a constant q. If the process responsible for the quenching influences only scintillation and does not influence charge collection at all, which is different from our assumption, the data may, for example, lie on a dot-dashed line [replacing $q^{-1}Q$ with Q in Eq. (2)]. As shown in the figure, this is not the case. S/S_0 for α particles has a maximum at $E \sim 1.5$ kV/cm and then falls on a straight line given by Eq. (3) with a constant q value (0.76 and 0.78 for 5.305- and 6.12-MeV α particles, respectively, which are 0.05 larger than those at zero electric field). This shows q increases with electric field at relatively low electric fields and then q becomes independent of the electric field at higher values, and it also shows that about 5% of the total charge is recovered by the electric field. The important implication of the result is that the quenching also influences charge collection. The maximum charge collection expected for infinite electric field for α particles can be as low as $0.78Q_\infty$ for a 6.12-MeV α particle if q stays constant for higher electric field, since the maximum charge collection is N_i $(= Q_\infty)$ if $q = 1$ and qN_i if $q < 1$. It is still possible that q increases again at a very high electric field and eventually all the charge produced is collected.

The result for fission fragments is also shown in Fig. 2. The charge collection and the change in scintillation for fission fragments were too small to see whether Eq. (3) applies to fission fragments.

The stopping power changes significantly along an α-particle track; nevertheless, the whole range of the track is influenced by quenching, since the scintillation efficiencies are almost the same for 5.305- and 6.12-MeV α particles.[8]

The increase of the scintillation yield, and that of the quenching factor q, at low electric fields, may be due to a contribution from a part which is lost in the quenching in the absence of electric field. Under an electric field, the distributions of positive and negative charges are changed, and some electrons are removed by the electric field.

Then the density of excitons and electron-hole pairs, and as a result the quenching, is decreased. Present results show, surprisingly, that this effect takes place only at low electric fields.

This increase was not observed for fission fragments. Since the range and velocity for fission fragments are comparable to those for α particles, the excitation density for fission fragments will be more than 20 times that of α particles. Due to this unusual excitation density, only less than 3% of the charge produced by fission fragments was collected for electric fields as high as 38 kV/cm, and it was only about 0.2% at $E=1.5$ kV/cm where scintillation due to α particles has its maximum. Therefore, it is not strange that an increase in scintillation yield was not observed for fission fragments. Since the fraction of charge collected for fission fragments is so small, it is too early to conclude if q depends on the electric field or not.

One possible explanation of the effects of the electric field observed in the present experiment is that due to the track structure; the core and its surrounding part are fairly distinguished and the electric field is not expected to penetrate into the core. If the observations are only due to the track structure, it is difficult to explain why the scintillation increases only at a low electric field. One may divide an α-particle track into three parts: (1) a surrounding part where no quenching occurs, (2) a hard-core part where quenching occurs and the electric field cannot penetrate, and (3) a part where quenching occurs but the electric field can also penetrate. Then one may attribute the third part to the end of the α-particle track, and/or consider the effect of the electric field on this part including differences in angles of α tracks with respect to the electric field.

Another explanation is that the quenching takes place in a fairly short time. When the charge distribution is changed and/or some electrons are removed by the electric field within this time, it influences the quenching process and then q becomes large. However, after this short period of time it becomes difficult to influence the quenching process even with a very high electric field. The drift velocity of electrons in liquid argon becomes only twice as large when the electric field increases from 1.5 to 10 kV/cm ($\sim 4\times 10^5$ V/cm at $E=10$ kV/cm).[9] This may explain why q increases at a low electric field then stays constant at a high electric field. The self-trapping of the hole may also be important since it influences the distribution of positive ions and the recombination process. The self-trapping time of a hole is considered to be short ($\sim 10^{-12}$ s).[10] The time dependence of scintillation from liquid argon due to α particles and fission fragments shows only decay components of self-trapped excitons (~ 7 ns and 1.6 μs),[11] and shows that the quenching takes place within a short time, possibly the same order of magnitude as the self-trapping times of free excitons and holes. More studies are needed to explain clearly the effect of the electric field on the quenching process.

There is a controversy on whether recombination in the track of an α particle in liquid argon supports the geminate theory.[12-14] However, the quenching effect was not taken into account in those discussions.

The authors would like to thank Professor J. Kikuchi, Dr. E. Shibamura, and Dr. K. Masuda for many helpful discussions. We wish to acknowledge the participation of Dr. S. Suzuki in some of the preliminary experiments and the assistance of Mr. T. Sano in the experiments. We thank Dr. T. A. King for reading the manuscript.

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