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Critical test of geminate recombination in liquid argon

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Recombination and impurity attachment in liquid argon have been separated using a gridded ionization detector with adjustable cathode to grid distance. Recombination effects with an electron source and an α source were studied in the range 0.07 to 10 kV/cm. In contradiction to other recent results, our data do not support geminate recombination theory.

Ionization recombination in liquid argon has been studied many times with a variety of techniques.[1–5] Theories used to interpret the data are as follows: columnar theory[6,7] which treats the ionization track as an essentially immobile core of positive ions surrounded by a cylindrical distribution of electrons, and geminate theory[8,9] which treats each electron-ion pair as independent, moving only under the influence of their mutual attraction, the applied field, and diffusion. Despite the existence of data that did not fully support[1,2] columnar theory, it has been generally assumed and used to extract W_l, the ionization energy per ion pair, for liquid argon.[3,10–13] Subsequently, Fuochi and Freeman,[4] and Gruhn and Edmiston[5] presented data on recombination using a γ and an α source, respectively, that supported geminate theory. The effect on extracting W_l due to the use of a different recombination theory is expected to be small (<1 eV).[13] However, if one focuses on the difference of W_l for the liquid (23.6 ± 0.3 eV)[12] and W_g for the gas (26.4 eV)[14,15] to develop some understanding of the behavior of free electrons in liquid argon, then this uncertainty becomes more significant. In any case, a better understanding of recombination would be useful,[16,17] for example, in interpreting information from liquid ionization detectors, e.g., liquid argon shower counters,[18–20] liquid argon time projection chambers (TPC),[21–23] etc.

The critical test of geminate theory occurs at low fields (<1 kV/cm in liquid argon), where a definite field dependence is predicted, whereas the data of Fuochi and Freeman,[4] and Gruhn and Edmiston[5] are taken at relatively high fields.

Reliable recombination data at low fields have not been collected previously with the pulse ionization chamber technique because of the inability to separate the effects of recombination and impurity attachment. This latter becomes progressively more important in the low-field region.[19,24] Since impurity attachment alone is a function of drift distance, the effects of recombination and impurity attachment can be separated by measuring the collected charge as a function of drift distance.[25,26]

The liquid-argon gridded ionization detector, used for the present study, allows the cathode to grid distance, i.e., the ionization drift distance, to be adjusted in the range 0.5 to 3.5 cm,[27] and was used earlier to demonstrate long-distance drifting of ionization electrons in liquid argon, which is necessary for the development of the liquid-argon TPC. An attenuation length of 170 cm at 1 kV/cm has been achieved recently with a larger system.[28,29]

A ^{113}Sn internal conversion electron source (364 keV, 0.2 μCi) or an ^{241}Am α source (5.64 MeV, 0.02 μCi) could be inserted in the cathode. Source activities and diameter (1.5 cm) were chosen to minimize bulk recombination and space-charge effects due to the accumulation of positive ions.[30] To test the consistency of our data, a 0.02-μCi ^{113}Sn source was also used.

To ensure total grid transmission, the ratio of the collecting field between the grid and anode (E_C) and the drift field between the cathode and grid (E_D) is always kept greater than 2. The collected charge was measured using standard commercial electronics. The amplifier shaping-filter time constant used is typically 2 μs. A time constant of 1 μs was used to confirm that electronics effects were insignificant at the low drift fields where electron drift velocity is low.

Data were collected using batches of liquid argon containing different types and amounts of impurities. For the electron source (^{113}Sn), data were collected using liquid argon with 3- and 6-ppb contamination (oxygen equivalent). After correcting for attachment, the data from the two sets were mutually consistent.

For the α source (^{241}Am), data were collected with a batch of liquid argon that deteriorated steadily because of a very small LN_2 leak from the cooling jacket. Initially no attachment was observed, though the collected charge decreased measurably with time (a few days). Subsequently, attachment was observed. After correcting for attachment, the collected charge at a fixed drift field was lower still. However, the fractional decrease in the collected charge was a constant, independent of the drift field. This effect due to N_2 impurity had been reported[24,25] at higher fields.

The recombination data, corrected for impurity attachment, are shown in Figs. 1 and 2 for the electron and the α source, respectively. With the batches of liquid argon used, attenuation lengths were measurable at low drift fields (<0.5 kV/cm). A linear dependence of the attenuation length on drift field provides a reasonable fit in this field region and it is extrapolated directly into the high-field region where attachment was too small to be measured. Therefore, corrections to the collected charge from impurity attachment were small in the high-field region (<2% at 2 kV/cm), but were significant at low fields (~20% at 0.20 kV/cm for the longest drift distance used).

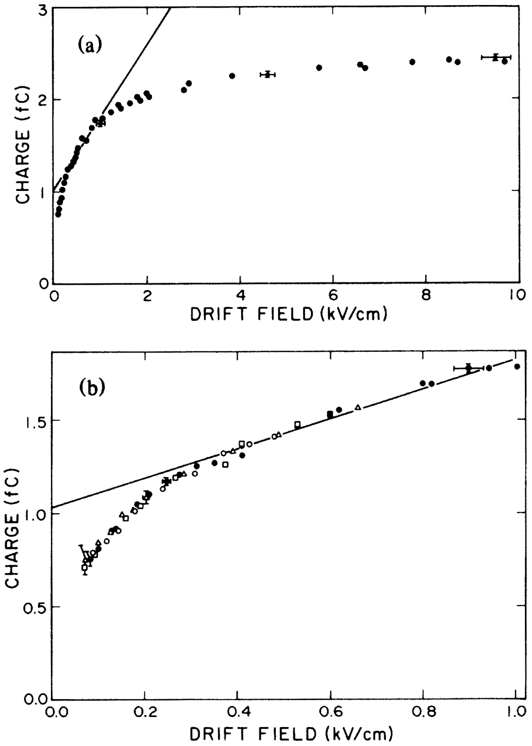

FIG. 1. The free-ionization electron charge collected using a ^{113}Sn source is shown vs drift field for the high-field region (a), and the low-field region (b). The different symbols represent data from different drift distances compensated for the effects of impurity attachment. The error in the collected charge is caused by uncertainties in determining the ^{113}Sn peak (± 0.02 fC), and measuring the attenuation length. The error in the field is caused by uncertainties in measuring the cathode-grid separation. The straight line is the best fit of geminate theory to the data.

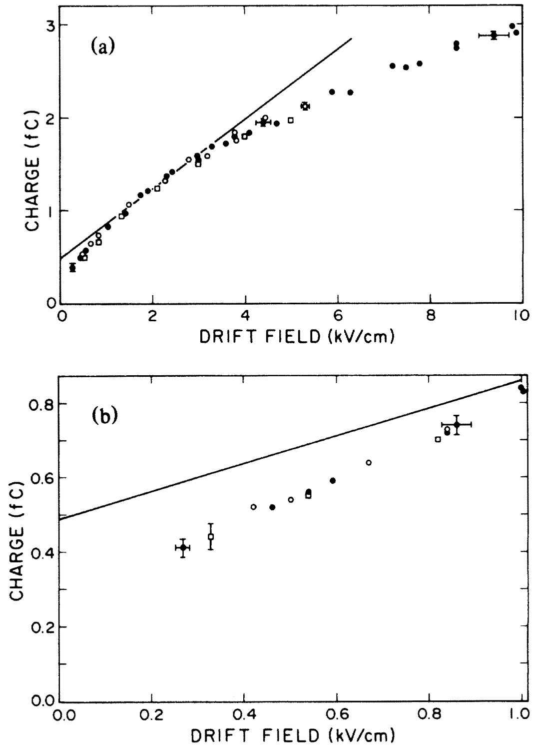

FIG. 2. Data corresponding to Fig. 1, but using an ^{241}Am source.

For an independent check on the pulse-ionization chamber approach above, we have measured the anode current with an electrometer using the electron source at the cathode. This approach gives the total charge escaping recombination directly irrespective of attachment to impurities.[24,25] However, γ's from the ^{113}Sn source deposit ionization over a large volume including the region between the grid and anode.[31] This can give rise to a constant residual current for a fixed grid voltage even if E_D is reversed. These data are shown in Fig. 3. After subtracting this residual current, we find that the ratio of the current data to the charge data remains constant within errors (about 2.5%) over the E_D range 0.2 to 4 kV/cm. This ratio is also shown in Fig. 3.

Our data can also be compared directly with those

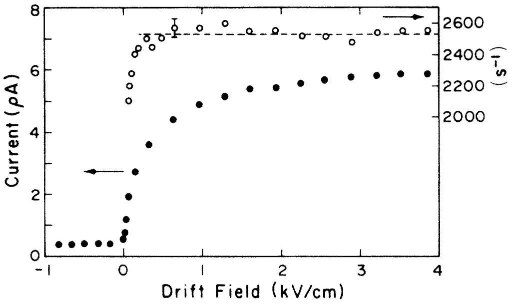

FIG. 3. Closed circles show the measured anode current vs drift field using a 0.2-μCi ^{113}Sn source. Note the existence of a residual current with reversed drift field. Measurement errors are ±1.5%. Open circles show the ratio of the measured current, with residual current subtracted, to the measured charge. This ratio is constant within errors (2.5%) between 0.2 and 4 kV/cm.

reported earlier at high fields where impurity attachment is less serious. For electron sources, the equation

$$Q(E_D)=Q_0/(1+k/E_D) \tag{1}$$

predicted by columnar theory has been used to fit the saturation curve at high fields.[12,26] Using Eqs. (1) in a $1/Q$-vs-$1/E_D$ fit for the ^{113}Sn data with E_D greater than 2.8 kV/cm, we get $Q_0=2.55\pm0.04$ fC and $k=0.53\pm0.04$ kV/cm. This result is in good agreement with Shibamura *et al.*[12]

For the α source, existing data do not generally agree in the field region less than 10 kV/cm.[32] If we ignore overall normalization and examine only field dependence of the collected charge, our data have the minimum slope. Since only our data have been demonstrated to be free of impurity attachment, we believe that impurity attachment affected the earlier data.

Geminate theory predicts a specific dependence of the free-electron yield as a function of the drift field which is

$$Q(E_D)=Q_0[\exp(-r_{kT}/r_0)](1+E_D/E_{kT}) \ , \tag{2}$$

for $E_D << E_{kT}$, where Q_0 is the free-electron yield at high fields, $r_{kT}=e^2/\epsilon kT$ is the Onsager length, r_0 is the thermalization length, E_D is the applied drift field, and $E_{kT}^{-1}=e^3/2\epsilon k^2T^2$ is the slope to intercept ratio predicted by geminate theory. At $T=91$ K and $\epsilon=1.53$,[33] $E_{kT}=1.31$ kV/cm. The solid line in Figs. 1 and 2 is the geminate theory prediction that best fits our data. The strong disagreement with geminate theory is evident at low fields.

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[30] In contrast to Ref. 5, we did not observe any decrease in the collected charge as a function of time after application of the drift field.

[31] For the ^{113}Sn source many more γ's than electrons are emitted (e_k/γ ratio is 0.44). Inhomogeneity of the drift field at the periphery of the detector has a substantial effect on recombination and collection of ionization induced by these γ's, particularly at low drift fields. Thus the current measurements are less reliable in this region. Note that the current measurement disagrees even more with geminate theory than the charge measurement.

[32] We examined data from Gruhn and Edmiston, Ref. 5; Willis and Radeka, Ref. 18; Hofmann *et al.*, Ref. 19; and Huffman *et al.*, IEEE Trans. Nucl. Sci. 26, 64 (1979).

[33] To within experimental uncertainty, no temperature dependence of the collected charge was observed when the pressure above the liquid N_2 bath was set between 35 and 45 psi (gauge).

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