Recombination of electron-ion pairs in liquid argon and liquid xenon

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We have measured the electric field dependence of electron-ion recombination in liquid argon and xenon. The observed relationship is incompatible with Onsager's geminate theory [Phys. Rev. **54**, 554 (1938)] of recombination so we have developed a new, single-parameter model to describe the data. The model is based on realistic assumptions about liquid argon and xenon and yields a simple, analytic result. This work is part of a program to build a high-resolution liquid-xenon detector.

I. INTRODUCTION

We are developing a liquid-xenon ionization detector for studying nuclear double β decay.[1] In order to detect this effect, it is desirable to achieve at least 0.5% resolution at the 2.5-MeV endpoint of the decay.[2] Better resolution than this should be possible. Several analyses[3,4] of fluctuations in electron yield suggest that liquid noble gas detectors are capable of 3 keV resolution at 1 MeV. However, the best resolution that has been observed at 1 MeV is 3.4% in argon and 6% in xenon.[3,5,6] It appears that the resolution is limited by some process other than Poisson fluctuations.

The thermodynamic and electrical properties of argon and xenon are well understood.[7–9] Initial electron-ion recombination has not previously been understood and does affect the resolution of an ion chamber. Figure 1 shows a typical fit of the commonly cited Onsager theory[10] to the total charge collected in a liquid-argon chamber irradiated by a ^{113}Sn source.[11] The purpose of this paper is to provide an alternative to the Onsager theory by demonstrating that recombination in liquid rare gases is a simple classical process and that Poisson fluctuations in the total collected charge do not affect the observed resolution except at very low electric field strengths (<1 kV/cm).

II. EXPERIMENTAL METHODS AND DATA

We have measured the electric field dependence of recombination in liquid argon and liquid xenon. Our detector is a simple ionization chamber[2] with an 85% transmission Frisch grid. The active volume has a 1-cm drift length and the signal is collected on a segmented anode with a central area of 1 cm^2. The measurements were made using a ^{113}Sn source that had been diffused into the cathode. The total activity of the source was $\sim$400 counts per second. The charge collection efficiency of the detector was calibrated ($\pm 5\%$) by observing the source with a cooled Si(Li) detector and assuming a pair energy of 3.60 eV. Figure 2 shows collected charge versus electric field in liquid argon and xenon. The relative errors are smaller than the data points.

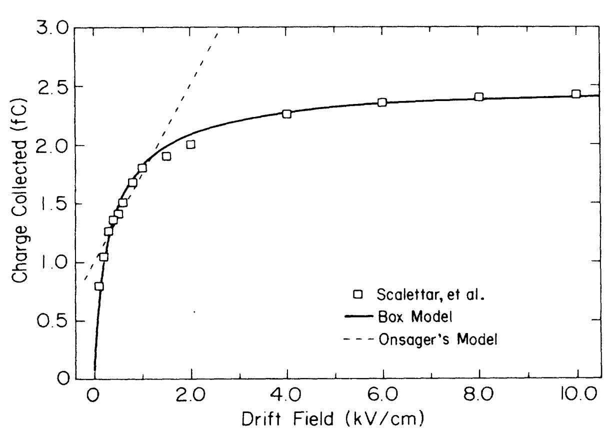

FIG. 1. The total charge collected as a function of electric field shows the effect of recombination in liquid argon after irradiation by electrons from a ^{113}Sn source. The data are from Scalettar *et al.* (Ref. 11). Onsager's model's best fit at low fields is shown here. The box model (this work) yields a better description of the data with $\xi E = 0.84$ kV/cm.

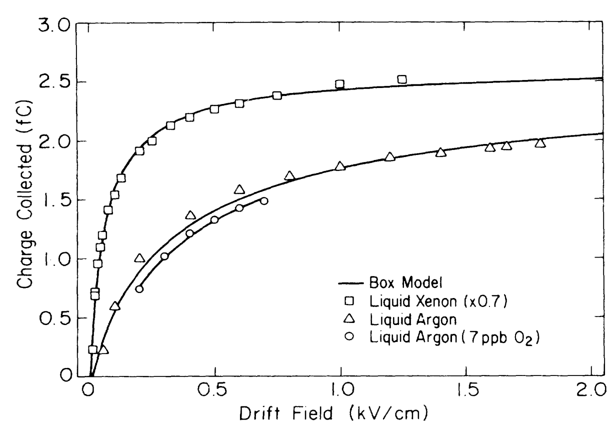

FIG. 2. A comparison between the total charge collected as a function of electric field in liquid argon and liquid xenon as measured in our apparatus. These fits include the effect of impurities as described in the text. The contaminant levels were less than 2 ppb with $\xi E = 0.84$ kV/cm in argon and $\xi E = 0.15$ kV/cm in xenon. The curve furthest to the right indicates the effects of 7 ppb of oxygen in liquid argon with $\xi E = 0.84$ kV/cm.

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III. DISCUSSION

The Onsager theory makes two fundamental assumptions about ion-electron recombination. The first is that each ion-electron pair is spatially separated (geminate theory). This appears to be appropriate because along the primary electron track, in liquid argon, the secondary electron interaction sites are separated by an average distance of ~ 100 atomic spacings. The second assumption is that the electron and ion interact via an infinite-range Coulomb force. But in argon, and especially in xenon, the high coefficient of polarization causes the induced dipole moments to reduce the effective charge of an ion within a few atomic spacings.[8] The resulting polarization potential falls off more rapidly than $1/r$ and the potential well around an ion is so deep that the ion travels by phonon-assisted tunnelling.[8,12] Consequently the ion mobility is several orders of magnitude smaller than the electron mobility.[13]

The defects in the Onsager theory prompted us to look for another description of the recombination process. Elegant models exist. The Smoluchowski equations form a basis for understanding the diffusion of charged particles with or without an external electric field[14,15] but suffer from the same defects as the Onsager theory. The more general problem of an electron travelling across a three-dimensional lattice with realistic interactions has been solved using a Green's function technique.[16] Unfortunately, the numerical solution of this problem is difficult and requires several parameters that have no direct relationship to experimental observables.

A simpler approach is to use a diffusion equation but to neglect the coulomb forces entirely and include recombination via a term that reflects the assumption that the rate of recombination depends on the density of the ions and electrons separately. Jaffe[17] discussed this model in 1913. The model is

$$\frac{\partial N_+}{\partial t} = -u_+ \mathbf{E}\cdot\nabla N_+ + d_+ \nabla^2 N_+ - \alpha N_- N_+ , \tag{1a}$$

$$\frac{\partial N_-}{\partial t} = u_- \mathbf{E}\cdot\nabla N_- + d_- \nabla^2 N_- - \alpha N_+ N_- , \tag{1b}$$

where N_+ and N_- are the ion and electron charge distributions, u_+ and u_- are the mobilities, d_+, d_-, and α are coefficients corresponding to the diffusion and recombination terms, respectively, and $\mathbf{E}$ is the external electric field. Jaffe attempted to solve this model by including the recombination term as a perturbation with the boundary condition that the initial distribution is a column of charge around the primary track.

Kramers[18] pointed out that in an external field the diffusion term is smaller than the drift or recombination terms and so a perturbative solution is unreliable. He solved the same equations by ignoring the diffusion term, including the columnar boundary conditions and assuming that the electron and ion mobilities are equal. Diffusion was added later as a perturbation.

In liquid argon and xenon, the diffusion term is very small. The electron diffusion rate is of the order of millimeters per meter of drift.[19,20] In addition, the ion drift velocity is three to five orders of magnitude smaller than the electron drift velocity.[9,13,21] Thus the diffusion terms in Eqs. (1a) and (1b) can be dropped, and the positive ion mobility set to zero. This simplifies the equations which, with a constant electric field applied along the z axis, become

$$\frac{\partial N_+}{\partial t} = -\alpha N_- N_+ , \tag{2a}$$

$$\frac{\partial N_-}{\partial t} = u_- E \frac{\partial N_-}{\partial z} - \alpha N_+ N_- . \tag{2b}$$

These equations can be solved exactly subject to the realistic boundary condition that each electron-ion pair is isolated. Equation (2a) can be substituted into Eq. (2b) and integrated over time to yield

$$\frac{\partial \ln N_+}{\partial t} = u_- E \frac{\partial}{\partial z}\left[\ln\frac{N_+(t)}{N_+(0)}\right] - \alpha N_+(t)$$

where we have used the initial condition $N_+(t=0) = N_-(t=0)$. To further simplify this equation, we introduce the definition $Y \equiv N_+(0)/N_+(t)$ and the variable transformation: $v = t - z/(u_- E)$ and $w = t + z/(u_- E)$. Applying the boundary condition $Y=1$ at $t=0$ ($v=-w$), the resulting equation is

$$\frac{\partial Y}{\partial v} = \frac{\alpha}{2} N_+(0) .$$

Next we apply the box model boundary conditions: the ion-electron pairs are isolated and the initial distribution of ions and electrons uniformly populates a box of dimension a (i.e., the box contains N_0 units of each charge at time $t = 0$.) Performing the integration over v (while holding w constant), letting $t \rightarrow \infty$, and integrating over all space, yields

$$\frac{Q}{Q_0} = \frac{1}{\xi}\ln(1+\xi), \quad \xi = \frac{N_0 \alpha}{4a^2 u_- E} , \tag{3}$$

where Q/Q_0 is the fraction of charge collected, and ξ is the single parameter upon which this theory depends. Thus, $\xi \rightarrow 0$ for perfect charge collection, and $\xi \rightarrow \infty$ in the case of complete recombination.

Figure 1 shows a fit of Eq. (3) to the data of Scalettar *et al.* These data are particularly interesting because the effects of impurity attachment and recombination have been separated by using a gridded ionization detector with an adjustable cathode to grid distance of 0.5 to 3.5 cm. The exponential attenuation due to oxygen impurities has been removed from the data presented in Fig. 1. The fit requires $\xi E = 0.84$ kV/cm and is sensitive to the calibration of the charge collection. We assume that the calibration of these data is perfect and fit our argon data with $\xi E = 0.84$ kV/cm. Figure 2 shows the fit.

Our model also describes the recombination rates observed when an α particle passes through liquid argon. Two data sets[11,22] are shown in Fig. 3. The α-particle track requires a value of ξE 560 times larger than that of the electron track due to the higher rate of recombination.

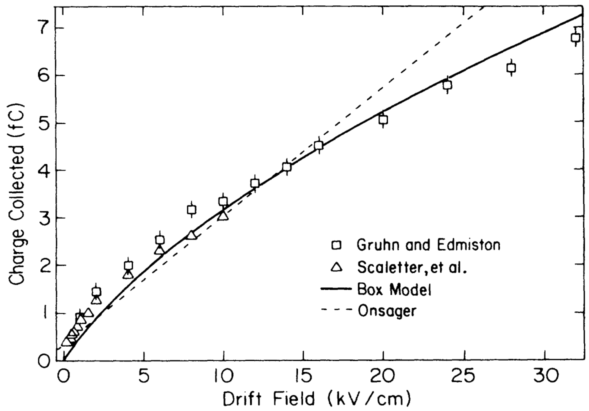

FIG. 3. Total charge collected as a function of electric field after an alpha particle passes through liquid argon. The solid curve is a fit to the combined data sets from Scalettar *et al.* (Ref. 11) and Gruen and Edmiston (Ref. 22) assuming no impurities and $\xi E = 470$ kV/cm.

Finally, our model of recombination can be used as a tool to measure impurity concentrations in liquid argon or xenon. The process of attenuation of electrons in noble gases by oxygen impurities occurs subsequent to initial recombination and has a characteristic exponential shape[23]

$$\frac{Q}{Q_0} = e^{-d/\lambda}, \quad \lambda = \beta\frac{E}{\rho} , \tag{4}$$

where d is the drift distance and β takes the value 0.15 in liquid argon.[23] (E is in kV/cm and the oxygen impurity concentration ρ is in ppm.) Multiplying the fraction of charge surviving recombination by the attenuation factor allows one to fit data with variable amounts of impurities. The data in Fig. 2 were fit using a point electron source and the effects of impurities. The best fit indicates that the total oxygen content was less than 2 ppb. For comparison, the data shown as circles in Fig. 2 were taken in a liquid-argon sample that was deliberately contaminated. The shift in charge collected corresponds to 7 ppb of oxygen.

IV. CONCLUSION

We have measured the electric field dependence of electron-ion initial recombination in liquid argon and xenon and we have developed a single-parameter model to describe it. The high quality of the fits to the data suggests that recombination is a simple classical process and that in the limit of infinite electric field 100% of the charge would be collected with an electron-ion pair creation energy of 23.6 eV in argon and 15.6 eV in xenon.[3,4] This means that Poisson fluctuations in the total collected charge (with or without a Fano factor) cannot be determining the resolution at fields greater than ~ 1 kV/cm since the observed resolution at high fields is a factor of 20 worse than predicted in argon and a factor of 75 worse than predicted in xenon at 1 MeV. This suggests that another process must be the cause of this discrepancy, and we are presently pursuing further experimental work to clarify this situation.

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