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FREE-ION YIELD IN LIQUID ARGON INDUCED BY ^{241}Am-α IRRADIATION

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Free-Ion Yield in Liquid Argon Induced by ^{241}Am-α Irradiation*

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Abstract

Recent measurement of free-ion yield in liquid argon by Gruhn and Edmiston (Phys. Rev. Lett. 40, 407, 1978) using ^{241}Am-α irradiation has been analyzed using a track model. Because of dense ionization recombination dominates in the core giving insignificant free ions. In the time scale of observation the δ-rays mostly degenerate into ion-pairs to which the Onsager probability is applicable. Electron thermalization distance is found to be 232 nm, the same as for γ radiolysis and in agreement with Freeman (Phys. Rev. B previous comment). Secondary electrons of energy about 200 eV can effectively penetrate the core.

In a recent work [1], Gruhn and Edmiston have determined the free-ion yield in liquid argon using ^{241}Am-α irradiation. The G-value, or 100-eV radiation yield, is found to be 0.046 in the limit of zero external electric field and it increases linearly with that field as predicted by the Onsager theory[2]. Freeman[3] criticizes several circumstantial and interpretive aspects of this work. Indeed in two respects the conclusion of Gruhn and Edmiston may be modified: (1) their work is the first free-ion yield measurement using a heavy ion, not the first observation of geminate recombination in liquid argon which was observed earlier [4]. (2) The electron thermalization length need not be 28 nm as stated. In fact it can be, and probably is, about the same as for γ-irradiation. A somewhat detailed track model can rationalize these observations with the Onsager model as shown in the following paragraphs. On the other hand, certain features of the work of Gruhn and Edmiston[1] are eminently well defended. These are: (1) Although a cylindrical track model is much better suited for α-irradiation, <u>in the time scale of measurement</u> it is an entirely reasonable hypothesis that most regions of dense ionization, including the track core, are fully neutralized leaving isolated ion-pairs few and far between. To these the Onsager theory[2] of geminate recombination can be applied. Mozumder and Magee[5] essentially did this while analyzing the work of Hummel et al.[6] in n-hexane using ^{37}Ar-irradiation. (2) Alternative model using explicit columnar recombination was actually tried by Gruhn and Edmiston[1]. They rejected this interpretation decisively on the basis that the line widths were too narrow by a factor of about 5 to fit the prediction of the model. (3) The low field data of Gruhn and Edmiston[1] fit the Onsager equation:

$$G(E)/G(o) = 1 + e^3E/2\,\varepsilon k^2T^2 \qquad (1)$$

within 5% with values of χ^2 as 0.92 and 1.18 for two sets of data. Since there are no adjustable parameters in Eq. 1, the agreement may be taken to be a strong evidence in favor of the Onsager model.

A current model of heavy-ion tracks in condensed matter consists of a cylindrical core surrounded by penetrating secondary electron (δ-rays)[7]. The core does not contribute significantly to G_{fi} since the ionization density and the axial fields are high. The δ-rays must have a minimum energy ε_o to effectively penetrate the core. Those having less energy are gradually engulfed by the core and indistinguishable from it. Thus only δ-rays of energy $\geq\varepsilon_o$ may contribute to G_{fi}. Ionization on δ-ray tracks are also more-or-less dense, meaning that the intervening ions quickly neutralize leaving those at the track extremities which behave like geminate pairs. It is believed that free-ions escaping from such geminate pairs are the only ones observed by Gruhn and Edmiston[1] in the μs time scale. A similar procedure was used by Mozumder and Magee[5] for ^3H and ^{37}Ar radiation-induced free-ion yield in hexane. We assume, as they did, that the geminate pairs do not interfere with each other; otherwise the Onsager theory prevails. Let S be the stopping power of the medium toward the α-particles and f the fraction of deposited energy contained in the core[7]. Assuming a Rutherford distribution in energy, the average number of δ-rays per unit track length is given by λ/ε^2 such that

$$(1-f)S = \lambda \ln(\varepsilon_m/\varepsilon_o). \qquad (2)$$

In Eq. 2 λ is a track constant and $\varepsilon_m = 2mv^2$ where m is electron mass and v is the α-particle velocity. From Eq. 2 the mean number of penetrating ($\varepsilon>\varepsilon_o$) secondary electrons in energy interval $d\varepsilon$ at ε is given by $(1-f)S(\ln\varepsilon_m/\varepsilon_o)^{-1}d\varepsilon/\varepsilon^2$. If $\varepsilon < 2\varepsilon_o$, then these electrons do not have any tertiaries that can contribute to the free-ion yield. For $2\varepsilon_o<\varepsilon<\varepsilon_m$ the mean number of energetic ($> \varepsilon_o$) tertiaries produced by a secondary of energy ε is given by $(\kappa-1)(\ln\kappa)^{-1}$ where $\kappa = \varepsilon/2\varepsilon_o$.[5] Since ε_m for a 5.3 Mev α from ^{241}Am-decay is 2.88 keV, the contribution of higher generation electrons may safely be ignored. Thus the total number of δ-rays produced per unit track length and contributing to G_{fi} is given by

$$n_\delta/L = \lambda\left[\int_{\varepsilon_o}^{2\varepsilon_o}\frac{d\varepsilon}{\varepsilon^2} + \int_{2\varepsilon_o}^{\varepsilon_m}\left\{1+\frac{(\kappa-1)}{\ell n\kappa}\right\}\frac{d\varepsilon}{\varepsilon^2}\right] = \frac{\lambda}{2\varepsilon_o}\left[1 + \int^{\varepsilon_m/2\varepsilon_o}\left(1+\frac{\kappa-1}{\ell n\kappa}\right)\frac{d\kappa}{\kappa^2}\right]. \quad (3)$$

There is considerable uncertainty regarding the range of low energy electrons in matter[8-11]. Fortunately in liquid argon, the thermalization length r_{th} is so large that the range obtained by integrating the electronic stopping power is always negligible. Thus, to a good approximation, the number of free-ions obtainable per unit track length is given from Eq. 3 as follows:

$$n_{fi}/L = \left(\frac{\lambda}{2\varepsilon_o}\right)\exp(-r_c/r_{th}) \left[1 + \int_1^{\varepsilon_m/2\varepsilon_o}\left(1+\frac{\kappa-1}{\ell n\kappa}\right)\frac{d\kappa}{\kappa^2}\right]. \qquad (4)$$

In Eq. 4 r_c is the Onsager length. Since S is the energy loss per unit track length, the 100-ev free-ion yield is given from Eqs. 2 and 4 as

$$G_{fi} = \frac{100\,(1-f)}{2\varepsilon_o \ln(\varepsilon_m/\varepsilon_o)} \exp(-r_c/r_{th}) \left[1 + \int_1^{\varepsilon_m/2\varepsilon_o} \left(1+ \frac{\kappa-1}{\ln\kappa}\right) \frac{d\kappa}{\kappa^2} \right]. \quad (5)$$

Equation (5) is only apparently independent of S; f depends on particle energy and is thus indirectly a function of S. The maxiumum transferable energy ε_m, of course, depends on particle velocity, but this is a minor dependence. Physically speaking, if S is not sufficiently large, a cylindrical core will not be obtained. In addition f is also a mild function of ε_o.

The Onsager length (r_c) in liquid argon is 127 nm. Taking the ionization yield and γ-induced free-ion yield as 4.5 and 2.6 respectively[4] the mean thermalizaion length (r_{th}) becomes 232 nm. With particles of energy 1.32 Mev/amμ, the track model of Mozumder et al.[7] gives f = 0.73. For uncertainties of low -energy electron penetration[8-11] sometimes a core radius about 1.nm is used in unit density material[12]. Considering the density of liquid argon (1.4 g/ml), we obtain, on the basis of rms penetration of Mozumder[11], ε_o = 140eV. We shall nevertheless treat ε_o as an adjustable parameter. Taking ε_m as 2.88 KeV, we have used Eq. 5 to calulate G_{fi} as a function of ε_o with f = 0.73 and the result is shown in Fig. 1. From this figure we obtain G_{fi} = 0.046 for ε_o= 200eV in agreement with experiment of Gruhn and Edmiston[1]. The value of f is expected to increase slightly with ε_o. Figure 2 shows the variation of G_{fi} with f at three values of ε_o. From such calculations we find that ε_o varies from 186 eV to 140 eV to 100 eV as f is varied in the important range 0.75 to 0.8 to 0.85 while keeping G_{fi} constant all along at 0.046.

In conclusion we may state that: (1) α-induced free-ion yield in liquid argon can be explained in terms of ion-pairs arising out of δ-rays from tracks. The external field effect is then normal as for γ-irradiation. (2) The thermalization length (232 nm) may be assumed to be the same as for γ-irradiation. (3) Secondary electrons of energy about 200 eV can effectively penetrate the core. Ranges of variation of ε_0 and f, to obtain agreement with experiment, are entirely consistent with track model.

Figure Captions

Fig. 1. Variation of calculated free-ion yield induced by ^{241}Am-α in liquid argon with ε_0, the energy of secondary electron needed to penetrate the core; f denotes the fraction of deposited energy to be found in the core.

Fig. 2. Dependence of calculated free-ion yield induced by ^{241}Am-α in liquid argon upon f, the fraction of energy deposited in the core, at three values of ε_0, the secondary electron energy needed to penetrate the core.

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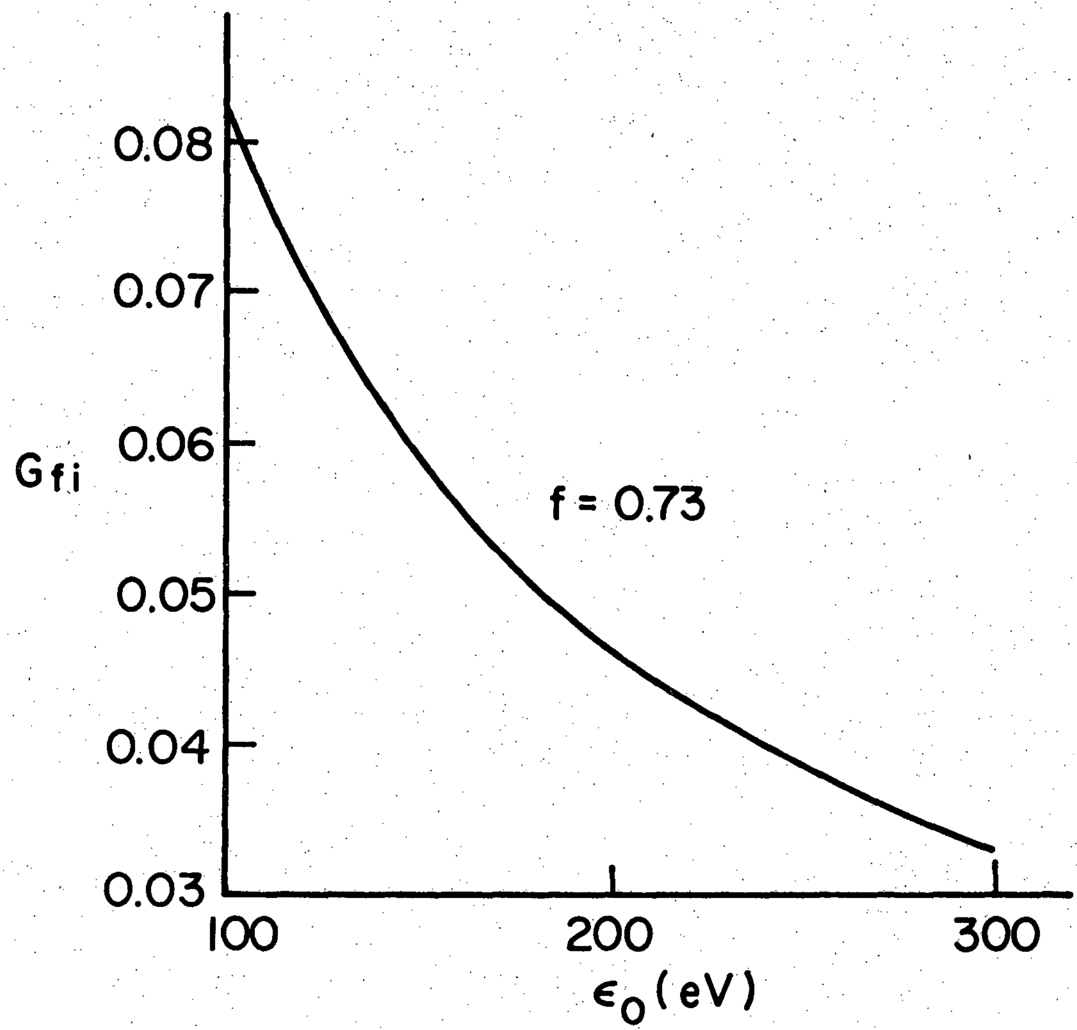

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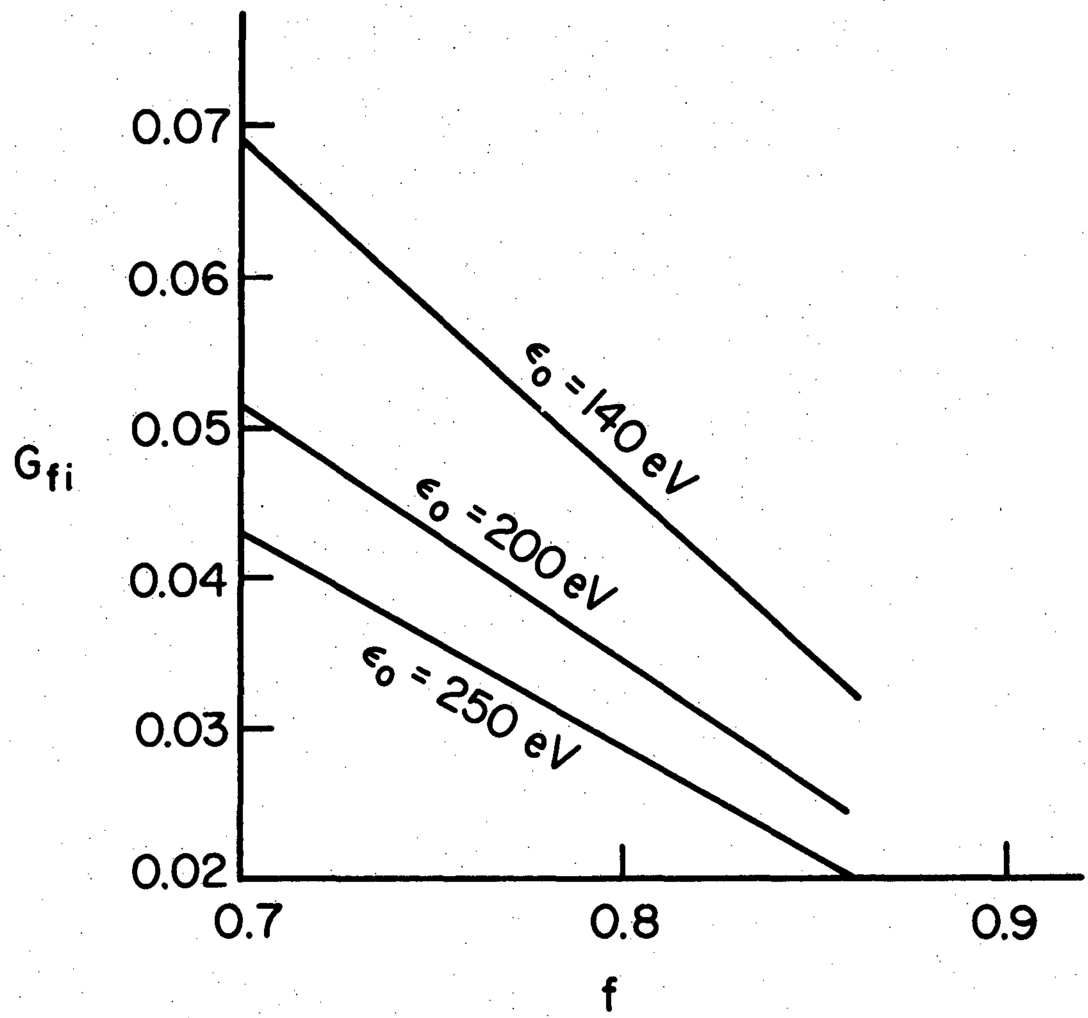
Fig. 1


0.07
0.06
0.05
0.04
0.03
0.02
G_{fi}
0.7
0.8
0.9
f
ϵ_o = 140 eV
ϵ_o = 200 eV
ϵ_o = 250 eV

XBL 793-8969

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