Luminescence quenching in liquid argon under charged-particle impact: Relative scintillation yield at different linear energy transfers

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A theoretical model is presented for the linear-energy-transfer (LET) variation of the relative scintillation yield in liquid argon. It is based on energy partition between the core and the penumbra of the charged particle track with little quenching in the penumbra except for fission fragments. Scintillation from the core can be quenched significantly by a biexcitonic mechanism. Some detailed calculations indicate that the electron-ion recombination may occur before exciton self-trapping. Fairly good agreement with experiment has been obtained with respect to the relative variation of the scintillation yield with LET using a diffusion-reaction model of free excitons with a specific reaction rate within acceptable limits. At the same LET different heavy-ion tracks can develop different quenching ratios depending on the density of deposited energy in the core.

I. INTRODUCTION

Absorption of ionizing radiation in liquid argon (LAr) produces no permanent chemical change. It is manifested in free charge carriers [mostly at low linear energy transfer (LET)], as scintillation from resultant self-trapped excitons or degraded as heat. The average number of electron-hole pairs produced by the absorption of energy T in LAr is given by $N_i = T/W$ where the W value has been measured to be 23.6 eV.[1] Additionally a certain number of excitons are directly produced with the ratio[2] $N_{ex}/N_i \doteq 0.21$. It is also known or conjectured on reasonable grounds that these excitons are quickly self-trapped (in $\sim 10^{-12}$ s) and the same applies to excitons produced by electron-hole recombination. This time is comparable to the hole self-trapping time.[3,4] The self-trapped excitons Ar_2^* give vacuum ultraviolet (vuv) scintillation. Before self-trapping the free excitons can diffuse and undergo biexcitonic quenching with a specific rate k accordingly as

$$X^* + X^* \rightarrow X + X + e\,(\text{kinetic energy}) \qquad (1)$$

when the excitation density is very high. The above reaction has been proposed as a possible mechanism of scintillation quenching.[5]

A series of experiments has been conducted to study the scintillation yield (the light intensity per unit absorbed energy) in liquid argon as a function of LET over more than four orders of magnitude,[6] using ionizing particles from radioactive sources and accelerators. Of the particles investigated, α particles, fission fragments, and relativistic Au ions show considerable quenching due to high excitation density. As we will show later about 30% of the energy deposited by α particles or relativistic Au ions is lost in quenching whereas only $\sim 17\%$ is available for luminescence by fission fragments.

It is important to note that although relativistic Fe and Kr ions, respectively, of 705 and 730 MeV/n energy, have much larger LET's than that of α particles, these do not show significant quenching. This fact demonstrates that quenching is not determined by LET alone, but also by the details of track structure, such as the radial distribution of energy deposition, which is given mainly by the velocity of the incident particle.

The $^1\Sigma_u^+$ and $^3\Sigma_u^+$ states of Ar_2^* give vuv scintillation. The emission lifetimes from these states have been observed to be the same for electrons, α particles, and fission fragments.[5] If the interactions between these low-lying molecular states were regarded as a possible quenching mechanism then the decay times would be LET dependent. The experimental fact that the decay times are indeed LET independent therefore rules out the participation of these states from the quenching process. The scintillation curves in Fig. 2 of Ref. 5 show a subnanosecond risetime for electron excitation but much faster risetime for α-particle and fission-fragment excitations. These observations suggest that the electrons and holes recombine under high LET excitation much faster than the electron thermalization time which is reported to be ~ 500 ps in solid Ar.[7]

We propose a quenching model based on energy partition between the core and the penumbra and a diffusion-reaction scheme of free excitons. A rapid recombination model due to a strong cylindrical field will also be discussed.

II. THEORETICAL FRAMEWORK

From the radiation physics point of view the structure of a heavy-ion track is conveniently described in terms of a core and a penumbra.[8] The core is the inner zone of relatively high-energy deposition density created by direct interaction with the primary particle and also to some extent by energy transfer from secondary electrons or δ rays. The penumbra, the outer region surrounding the core, is a zone of relatively low-energy deposition density produced by energy transfer from emergent secondary electrons. Figures 1(a), 1(b), and 1(c) show, respectively, the core and penumbra of tracks of α particles, fission fragments, and relativistic Au ions in LAr. Typical values of deposited energy density by a heavy ion in the core and penumbra are of the order of $\sim 10^{-1}$ eV/Å^3 and $\sim 10^{-4}$ eV/Å^3, respectively.[9] It is therefore clear that a biexcitonic quenching mechanism, such as depicted in Eq. (1), cannot be very effective in the penumbra region of ordinary heavy-ion tracks. In what follows we will assume that quenching occurs exclusively in the core except for fission fragments where due to high ionization density the quenching efficiency will be assumed to be equal in both the core and the penumbra. With the exception for fission fragments then the energy T_s available for scintillation should be given by

$$T_S = qT = q_c T_c + T_p , \quad T = T_c + T_p , \tag{2}$$

where T is the energy delivered into liquid argon with the components T_c and T_p invested, respectively, in the core and the penumbra, q is the overall observed quenching factor ($q = 1$ for no quenching)[6] and q_c is that factor attributable to the core. That is, q_c is the fraction of energy deposited in the core that survives quenching. [For fission fragments only the first of the equations (2) can be used.] For example, if we take $q = 0.71$ for 5.31-MeV α particles in LAr and $T_c/T = 0.72$ (see Sec. II A), then Eq. (2) implies $q_c = 0.6$, or that 40% of the energy deposited in the core is quenched in regard to scintillation. Similar estimations can be made for other primary ionizing particles.

Our theoretical model for calculating q as a function of particle LET and velocity can be subdivided into three parts. First, we compute the deposited energy partition between the core and the penumbra, i.e., compute T_c/T. Second, we develop a theory for electron-ion recombination on a track in LAr and strive to show that most recombination is indeed completed within the time scale of exciton self-trapping (see the Introduction). Finally, we employ a diffusion-kinetic scheme to calculate the efficiency of biexcitonic quenching [cf. Eq. (1)] during the time scale of self-trapping of the free exciton.

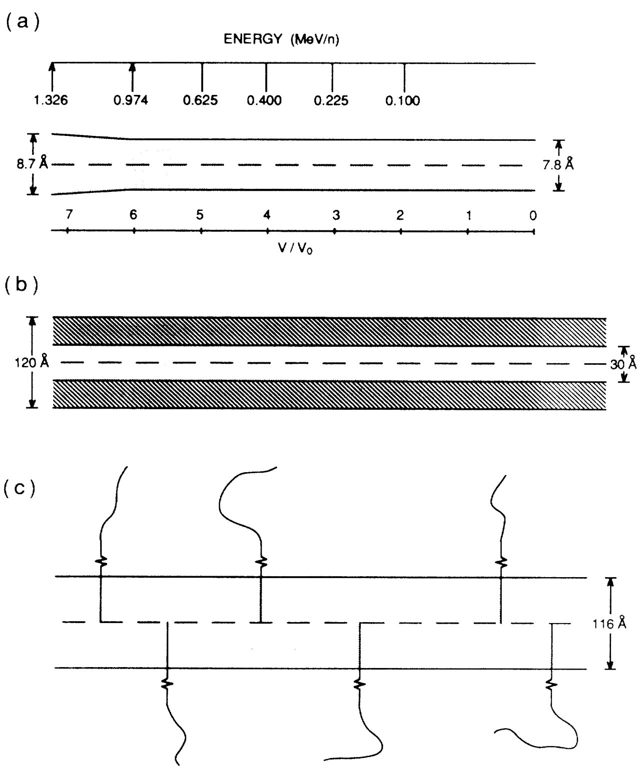

FIG. 1. The core and penumbra structure of heavy-ion tracks in LAr: (a) α particles; (b) fission fragments; (c) Au ion (870 MeV/n). The core is shown as dotted. For α particles the penumbra is a disjointed collection of emergent δ rays (not shown). These are ~200–2000 eV in energy, ejected nearly perpendicular to the track axis and have LET's somewhat less than that of the main particle. In the case of fission fragments the δ rays coalesce to form a quasicontinuous penumbra although the energy density is less than that in the core. The average δ ray on the Au ion track has energy ~2 keV; these are ejected nearly perpendicular to the track axis with negligible stopping or scattering within the core. The mean ranges of these δ rays are ~2000 Å, their LET's are orders of magnitude smaller than the main particle and they overlap little. The wavy lines indicate breaks in the path of δ rays. See text for details.

A. Energy partition

For nonrelativistic ions it has been shown in Ref. 8 that the core radius is given by Bohr's impulse principle, i.e., by the maximum impact parameter r_0 capable of exciting the lowest electronic state E_1 of the medium. Thus $r_0 = hv/2E_1$ where h = Planck's constant divided by 2π and v is the incident ion velocity (*vide* infra for some modification of this concept). The energy deposition within the core is computed by adding the energy deposited directly by the main particle and the amount transferred from secondary electrons (δ rays) while penetrating the core. The calculations of Ref. 8 was actually made for water. However, the stopping powers of water and LAr are very similar, the higher density of LAr almost exactly compensates for its higher value of the mean excitation potential. Thus the results of Ref. 8 can be used safely for LAr as well. The nonrelativistic particles used in the experiments[6] are ^{210}Po-α (1.33 MeV/n), ^{212}Bi-α (1.51 MeV/n), ^{212}Po-α (2.20 MeV/n), ^{252}Cf-α (1.53 MeV/n), ^{252}Cf-*ff* (0.98 MeV/n-light and 0.56 MeV/n-heavy). Since the relativistic correction to stopping power in the lowest order varies as β^4 where $\beta = v/c$, protons of 17.8 and 38.2 MeV ($\beta = 0.192$ and 0.277) should also be considered nonrelativistic. According to Ref. 8 the fractional energy deposited in the core (T_c/T) for the above sequence of particles is given, respectively, by 0.72, 0.71, 0.69, 0.71, 0.76, 0.80, 0.75, and 0.75. The last two numbers for the proton tracks actually lie outside the range of Fig. 7 of Ref. 8. However, a smooth extrapolation in this asymptotic region is quite reliable.

The relativistic ions used in the experiments[6] are p (1.04 GeV), He (1.04 GeV/n), Ne (631 MeV/n), Ne (1.35 GeV/n), Fe (705 MeV/n), Kr (730 MeV/n), La (1.08 GeV/n), and Au (870 MeV/n). For such ions the density effect becomes important and we therefore use Fermi's[10] theory of stopping power. It has been shown[11] that for relativistic particles the core radius is given by $r_0=\lambda\beta$ where λ is the maximum core size. This is obtained by equating the radial outflow of energy at r_0 according to Fermi's theory to the neglected part of the stopping power by application of Bohr's impulse principle, implying that direct deposition of energy by the main particle beyond r_0 is negligible. The quantity λ is given by $x_{\max}c/\omega_0\epsilon^{1/2}$ where $x_{\max}=1.074$, ω_0 is a typical excitation energy, and ϵ is the dielectric constant. For LAr we take $\epsilon=1.56$ and $\hbar\omega_0=25$ eV and obtain $\lambda=68$ Å. For relativistic particles of the above list β changes little (0.80–0.91), reflecting a corresponding change in the core radius (54–62 Å). This may be contrasted with $r_0=4$ Å for α particles using the Bohr criterion with $E_1=12$ eV. The equipartition of deposited energy between glancing and knock-on collisions has been substantiated for relativistic particles.[12] Fano[13] gives the energy distribution of δ rays produced by knock-on collision as

$$dn_\delta/dT_\delta=\kappa NZ[1/T_\delta^2-(1-\beta^2)/2mc^2T_\delta] ,$$

where $\kappa=4\pi z^2e^4NZ/mv^2$ is the kinematic factor, ze is the incident particle charge, m is electron mass, N is the number density of the medium, and Z is the atomic number of the medium. The average energy of δ rays with respect to this distribution is then given by

$$\langle T_\delta\rangle=\frac{\ln(T_m/I)-\beta^2}{(I^{-1}-T_m^{-1})-(1-\beta^2)(2mc^2)^{-1}\ln(T_m/I)} , \quad (3)$$

where $T_m=2mv^2/(1-\beta^2)$ is the kinematically maximum value of T_δ and the lowest energy of δ rays is taken to be I, the mean excitation potential, in consistence with the equipartition principle. For LAr $I=200$ eV; for β between 0.80 and 0.91, T_m lies in the range 1.78–4.81 MeV, thus giving from Eq. (3), $\langle T_\delta\rangle$ between 1700 and 1852 eV. The ejection angles for δ rays with energy $\langle T_\delta\rangle$, given by $\cos^{-1}(\langle T_\delta\rangle/T_m)^{1/2}$ lie between 88.0° and 88.8°, i.e., almost perpendicular to the track axis. The stopping power of low-energy electrons has been calculated rather accurately for water by LaVerne and Mozumder.[14] Following a similar procedure we compute the range and stopping power of ~2 kV electrons in LAr, respectively, as 2143 A and 0.61 eV/A. The elastic scattering of δ rays is not expected to be significant within the core radius (~60 Å), neither is the variation of the stopping power within it. Thus the estimated energy loss of a typical δ ray within the core is ~60×0.61 or 36 eV. This is about 2% of the δ-ray energy or overall ~1% of the total energy loss. Thus the energy partition between core and penumbra for experimental relativistic particles[6] turns out to be 51:49, or for our purpose, roughly 50:50. In sharp contrast with nonrelativistic particles, this implies that very little energy of the δ rays is deposited in the core and that basically an equipartition holds between the core and the penumbra.

B. Recombination time scale

The current estimate of electron thermalization time in LAr is ~500 ps.[7] Although such a time may be relevant for high-energy electron tracks it is greatly in excess of the self-trapping time of free excitons which is variously estimated to be in the range ~1–10 ps.[3,4] Since the proposed mechanism of scintillation quenching favors that the electron-ion recombination be substantially over before the time scale of self-trapping we must conclude that such recombination actually occurs prior to thermalization. Indeed we will demonstrate that the electron, instead of being thermalized on a heavy-ion track, comes to a classical turning point provided by the field due to the line of positive charges on the track axis. Following this the electron is repeatedly drawn towards the track axis and at each pass it undergoes a small probability of recombination. This idea will be developed in the following paragraphs.

After ejection from the track axis some of the secondary electrons penetrate the core and others of less energy are essentially contained within it. Taking a Rutherford (E^{-2}) spectrum of ejected electrons between the ionization potential (14 eV) and an estimated upper limit (~200 eV) for penetration based on the core radius and range-energy relationship, the mean energy $\langle E\rangle$ is found to be 40 eV, for which the range and stopping times are computed as 28.6 Å and 1 fs.[14] Elastic-scattering mean free path L for low-energy electrons, between subexcitation energy E_1 (12 eV) and $\langle E\rangle$, has been estimated by Mozumder[15] ~3 Å and by Atrazhev and Yakubov[16] ~5 Å. Taking $L=4$ Å and using a random-walk model the rms penetration to subexcitation energy is computed as $R_1=4\,(28.6/4)^{1/2}$ or 11 Å. From here the electron executes random flight in the cylindrical field due to an axial density of P positive charge per unit length and comes to a classical turning point (CTP) at a radial distance R_{CTP} where its kinetic energy is reduced to zero. Then the electron is drawn back to the axis where it has some chance to recombine with one of the positive charges or to go back to the CTP, from where the sequence of steps is repeated until full recombination becomes possible. Figure 2 shows schematically one phase of such a motion where the zigzag course depicts the projection of the motion of the electron including random scattering.

In the cylindrical field due to a linear density of $+Pe$ charge on the track axis the distance of the classical turning point is given by

$$R_{\text{CTP}}=R_1\exp(\epsilon E_1/2Pe^2) ,$$

where we have ignored the small energy loss due to elastic scattering (at most ~2%) and equated the potential difference between R_1 and R_{CTP} with E_1. For 5.31-MeV α particles in LAr we compute $P=4.33\times10^7$ cm^{-1} from its range[17] and W value.[1] With $\epsilon=1.56$, $E_1=12$ eV, and $R_1=11$ Å (*vide* supra) we then estimate $R_{\text{CTP}}=50$ Å. The actual tortuous path length would be greater because of elastic scattering for which the mean free path Λ at

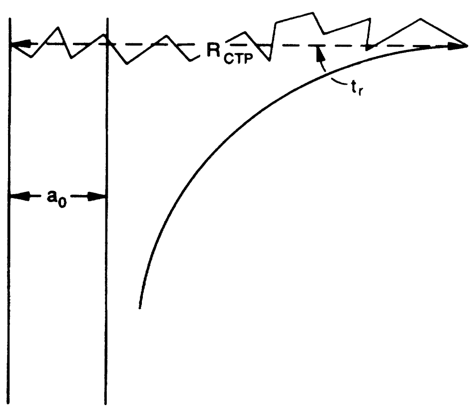

FIG. 2. Sketch of the random flight of an electron in the cylindrical field caused by the line of positive charges on track axis. $R_{\rm CTP}$ is the classical turning point and t_r is the return time from this to the recombination distance a_0.

middle energy (6 eV) has been estimated to be 10 Å.[15] The rms crooked pathlength is then $\sim R_{\rm CTP}^2/\Lambda$ or 250 Å in this example, giving the rms time to get to the CTP as ≈ 17.2 fs using an average velocity 1.45×10^8 cm s^{-1} corresponding to 6 eV kinetic energy. The return time t_r in the absence of scattering is simply calculated solving the radial equation of motion with the result

$$t_r/t_0 = \mathrm{erf}\{[\ln(R_{\rm CTP}/a_0)]^{1/2}\} ,$$

where a_o is the reaction radius, $t_0 = R_{\rm CTP}(\pi/2\alpha)^{1/2}$, and $\alpha = 2e^2/P\epsilon m$, m being electron mass. We take a_0 equal to the core radius (4 Å, *vide* supra), giving in the present example, $t_0 = 5.27$ fs and $t_r = 5.14$ fs. Similar to Bethe's[18] Umwegfaktor concept we assume the ratio of mean return time to that in the absence of scattering to be equal to the ratio of rms tortuous path length to the radial distance; i.e., $\langle T_r \rangle / t_r \approx (R_{\rm CTP} - a_0)/\Lambda = 4.6$. This gives $\langle T_r \rangle = 23.6$ fs. Since we are neglecting the small energy loss due to elastic scattering the time for outward and inward journeys should be statistically equivalent. We compute somewhat different times (17.2 and 23.6 fs) partly because of different procedures (thus reflecting the relative uncertainties of calculation) and partly because the starting (11 Å) and final (4 Å) distances are different. The total return time, apart from the small (~ 1 fs) time to slow down to subexcitation energy, may, however, be reliably estimated at (17.2+23.6) fs or ~ 40 fs. This example is for α tracks in LAr. For particles of higher LET, R_1 would be less but the energy differences will tend to remain the same, thus decreasing the time for a round trip pass through the CTP and the reaction radius. The number of passes required to complete recombination can be approximated as $k_D/k_{\rm expt}$ where k_D is the Debye rate and $k_{\rm expt}$ is the experimental specific rate of electron-ion recombination. This factor has been found experimentally to be[19] ~ 10, which then gives the recombination time scale on α tracks in LAr to be ~ 0.4 ps. Since the self-trapping time of excitons in LAr have been estimated to lie in the range 1–10 ps,[3,4,20] the above example shows that electron-ion recombination on heavy-ion tracks in LAr can essentially be completed before self-trapping.

C. Biexcitonic quenching kinetics

The relative scintillation yield in LAr has been thoroughly investigated as a function of LET over more than four orders of magnitude.[6] It is clear that this quantity reaches a peak for relativistic heavy ions of the Ne-La group (LET between 200 and 5000 MeV cm^2 g^{-1}) and it decreases on either side of this LET interval. Although some necessary details remain to be filled in, it appears that the loss of scintillation on the low-LET side can be explained in terms of electrons escaping recombination,[6] for which a probability 0.35 has been obtained for high-energy (~ 1 MeV) incident electrons.[21,22] We will therefore make no further comment on this aspect of the problem. On the other hand, the loss of scintillation at high LET was also observed by Salamon and Ahlen[23] for NaI(Tl) crystals. These authors proposed a second-order annihilation mechanism for scintillation quenching at high LET. Even though the exact quenching mechanism remained elusive a diffusion-reaction equation for the contributing species was set up and integrated in time until $\sim 10^{-8}$ s, which is the "trapped" exciton or *e-h* pair lifetime in this system. The computed result was compared with the experimental values for luminescence quenching. A similar second-order mechanism has been hinted at for luminescence quenching at high LET in LAr,[5,6] but no specific computation was made and no details of the mechanism was arrived at.

Any theory of scintillation quenching in LAr must contend with the fact that the observed light emissions are from self-trapped singlet and triplet excitons having the nature of molecular excimer states ($^1\Sigma_u^+$ and $^3\Sigma_u^+$). Furthermore the emission life-times are experimentally found to be independent of LET, i.e., the same under electron or α-particle excitation.[5] This rules out interactions between these low-lying excited molecular states as a possible quenching mechanism. To explain the experimental quenching results in LAr we propose a cylindrical track model including electron-ion recombination and diffusion-reaction of free excitons. The working hypothesis rests on the following: (1) Exciton formation, directly (with about 0.21 probability) or on ion recombination. (2) Competition between exciton diffusion and self-trapping. Biexcitonic quenching reduces scintillation in the cores of heavy ions and also in the penumbra of fission fragments. (3) Scintillation from self-trapped excitons. Since the self-trapped exciton lifetime is LET independent it is necessary that recombination must occur before self-trapping (this point has been established in Sec. II B).

In the biexcitonic quenching mechanism shown in Eq. (1), the electron carries the excess energy, which is, to a good approximation, $2E_{\rm ex} - E_g$, where $E_{\rm ex}$ is the excitation energy and E_g is the band gap. The electron loses this energy before recombination. One thus gets one excitation in place of two resulting in fewer phonon emission. The diffusion-kinetic equations for the free (index 1) and the self-trapped (index 2) excitons may be written as

$$\partial n_1/\partial t = D\nabla^2 n_1 - k n_1^2 - n_1/\tau - A_1 n_1 \tag{4}$$

and

$$\partial n_2/\partial t = n_1/\tau - A_2 n_2 , \tag{5}$$

where n is the exciton density, D is the diffusion coefficient of the free exciton, k is the specific rate of biexcitonic quenching, τ is the free exciton lifetime against self-trapping, and A_1 and A_2 are the respective radiative decay constants. The last two constants can be neglected since the free exciton emission is very weak[24] and self-trapped excitons are not responsible for quenching. Diffusion-reaction equations of the type of Eq. (4) are often encountered in radiation chemistry involving electrons, ions, and free radicals. One standard technique of solving this equation is by the application of the method of "prescribed diffusion." It means that the spatial dependence of the reactive intermediates (here excitons) is prescribed as a well-known mathematical function, often as a Gaussian which is the solution of the diffusion equation without reaction. The time-dependent normalization factor of the Gaussian function is then obtained by substituting the prescribed function into Eq. (4), followed by an integration. This kind of approximation is usually reliable when one is interested in the time dependence of the total amount of species (here excitons) rather than in their spatial variation. In cylindrical geometry one writes with Gaussian approximation[25]

$$n_1(r,t) = N(t)G(r,t) , \tag{6}$$

where $N(t)$ is number of free excitons per unit length at time t and

$$G(r,t) = (\pi a_t^2)^{-1}\exp(-r^2/a_t^2), \quad a_t^2 = a_0^2 + 4Dt \tag{7}$$

is a normalized distribution at any time t in the sense that $\int_0^\infty dr\, 2\pi r G(r,t) = 1$. Note that the Eq. (7) provides the "spread" of the spatial distribution in time by diffusion. Substituting Eqs. (6) and (7) in Eq. (4), integrating over all r and noting that $\int \nabla^2 G\, dv = 0$ by Green's theorem under standard boundary conditions, one gets

$$\begin{aligned}\frac{dN}{dt} &= -\frac{N}{\tau} kN^2 \int_0^\infty G^2(2\pi)dr \\ &= -\frac{N}{t} - \frac{kN^2}{2\pi(a_0^2 + 4Dt)} .\end{aligned} \tag{8}$$

Equation (8) can be linearized with the substitution $u = N^{-1}$ and solved in the usual manner. The result is

$$N(t) = N_0 e^{-t/\tau}\left[1 + \frac{kN_0}{2\pi}\int_0^t \frac{\exp(-t'/\tau)dt'}{(a_0^2 + 4Dt')}\right]^{-1} , \tag{9}$$

where N_0 is the initial number of excitons per unit length. The integral in Eq. (9) may be written as

$$\int_0^t \frac{\exp(-t/\tau')dt'}{(a_0^2 + 4Dt')} = \frac{\exp(a_0^2/4D\tau)}{4D}\left[E_1\left[\frac{a_0^2}{4D\tau}\right] - E_1\left[\frac{a_0^2 + 4Dt}{4D\tau}\right]\right] , \tag{10}$$

where $E_1(\xi) = \int_\xi^\infty \exp(-\xi)d\xi/\xi$ is the exponential integral for which extensive tabulation exists.

We observe the emission from the self-trapped excitons. The population of self-trapped excitons per unit length $N_2(t)$ is given by $N_2(t) = \int_0^\infty dr\, 2\pi r n_2(r,t)$ and from Eqs. (5)–(7) we obtain

$$\frac{dN_2(t)}{dt} = \frac{N(t)}{\tau} , \tag{11}$$

where we have ignored the radiative depopulation term $-A_2 n_2$ in Eq. (5) since the self-trapping time τ is much shorter than the lifetime of light emission. All the self-trapped excitons give vuv emission. Thus we have

$$N_2(t) = \int_0^t \frac{N(t')}{\tau} dt' . \tag{12}$$

The fraction q_c which survives biexcitonic quenching is now given by

$$q_c = N_2(\infty)/N_0 = \frac{\int_0^\infty [N(t)/\tau]dt}{N_0} , \tag{13}$$

which can be calculated using Eqs. (9) and (10) and an integration. The actual evaluation of the quenching factor q_c using realistic track constants (N_0, a_0), the free exciton diffusion coefficient (D), and rate parameters (k and τ) will be discussed in the next section.

III. RESULTS AND DISCUSSION

The constants needed for the quenching calculation are N_0, a_0, k, τ, and D. Here N_0 is the initial number of excitons in the core per unit length, produced either directly or on electron-hole recombination. For relativistic particles, the energy deposited in the sensitive region (Table 1 of Ref. 6) divided by the path length in the region (2 cm) was taken to be the LET and it was assumed that 51% of this was deposited in the core (Sec. II A). For α particles and fission fragments, the ranges were taken from Northcliffe and Schilling.[17] Although the LET changes considerably over the ranges of these particles it was assumed to be a constant at an average value along the track. The fraction of the energy deposited in the core is given in Sec. II A for those particles. For LAr

$W = 23.6$ eV and $N_{ex}/N_i = 0.21$ were used.

For the first calculations, it was assumed that the recombination time is much shorter than the free exciton lifetime τ. Therefore the radial distribution was taken to be a Gaussian with a size parameter the same as the core radius given by $\lambda\beta$ where $\lambda = 67.8$ Å (see Sec. II A) for relativistic ions. For some nonrelativistic ions such as α particles of energy less than 1 MeV/n, r_0 according to the Bohr criterion becomes smaller than the interatomic distance, in which case the latter (3.9 Å for LAr), is taken for r_0. The excitation density due to fission fragments is so large that it becomes unreasonably high if a core radius given by Bohr's impulse principle or an interatomic distance is taken for a_0. It probably implies that initially each Ar atom on the path of a fission fragment is multiply excited or ionized and very quickly the charges and excitations are shared by other atoms in the proximity. Therefore $r_0 = 16.5$ Å was determined such that the energy deposited is distributed in a single Gaussian, and the number density of excitons at $r = 0$ does not exceed that of Ar atom in LAr. The initial radial distributions determined in this way are shown in Fig. 3.

We can estimate the cross section σ for process (1) using a theoretical formula for Penning ionization in the gas phase.[26] Excitons are treated as particles moving rapidly in condensed media with a mass similar to the electron mass (effective mass $= 0.5m_e$ for solid Ar).[20] The rate constant k is given by $k = \sigma v$ where v is the thermal velocity of collision partners ($v \sim 1.2\times 10^7$ cm/s). Thus we have $\sigma \sim 2.5$ Å^2 and $k \sim 3\times 10^{-9}$ cm^3/s for dipole-dipole interaction, using the transition dipole moments for $Ar^* \to Ar$ and $Ar^* \to Ar^+ + e^-$ available in the literature.[27] This estimated value of σ may be a lower limit since the "free" exciton motion is so rapid that σ due to dipole-dipole mechanism is rather small.[20] The upper limit may be given by the hard-sphere cross section. We estimate a value for $Ar^* + Ar^*$ to be ~ 170 Å^2 using the hard-sphere radius of Ar^* as $r_{HS} = 7.4$ Å.[28] The free exciton lifetime is taken to be 1 psec and the diffusion constant $D = 1$ cm^2/s is used.[29]

The evolution of the quenching process is shown in Fig. 4 for 5.305-MeV α particles and for 870-MeV Au ions. The figure shows the ratios of the number of self-trapped excitons with and without quenching as a function of time (t) in unit of the free exciton lifetime τ. We have calculated the value up to $t/\tau = 4$. The limits show the values of q_c, which is the quenching factor in the track core [see Eq. (2)]. One quarter of the hard-sphere cross section was taken to compute the k value used in this figure. Comparing the quenching kinetics in the narrow, dense core of the α particle with that in the broad core of the Au ion ($r_0 = 58$ Å) we see from Fig. 4 that the quenching develops gradually for Au ions since in this case the core radius is comparable to the free exciton diffusion length, reported to be 100 Å for solid Ar.[29] For α particles a strong quenching occurs at a very early stage ($t/\tau \approx 0.01$) due to high initial density of excitons. The core then quickly diffuses out and quenching progresses slowly. The core quenching factors q_c obtained for α particles and Au ions with $\sigma = \sigma_{HS}/4$ are 0.65 and 0.45, respectively. The entire track q value is then obtained from Eq. (2) using T_c/T values of 0.72 and 0.51, respectively, for α particles and Au ions. These are shown in Fig. 5 together with the results for other particles and those obtained by using $\sigma_{d\text{-}d}$.

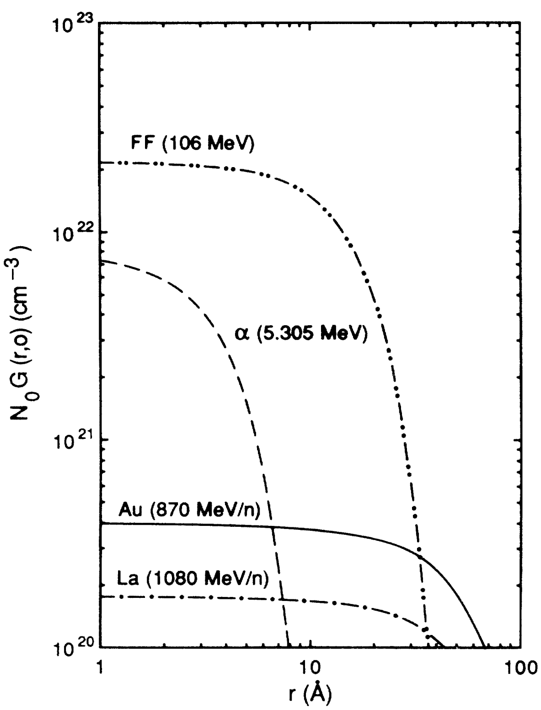

FIG. 3. The initial distribution of excited species, $N_0G(r,0) = [N_0/(\pi a_0^2)]\exp(-r^2/a_0^2)$, in the track core produced in liquid Argon by 5.305-MeV α particles (— — —), 106-MeV fission fragments (–··–··), 1080-MeV/n La (–·–·–), and 870-MeV/n Au ions (——).

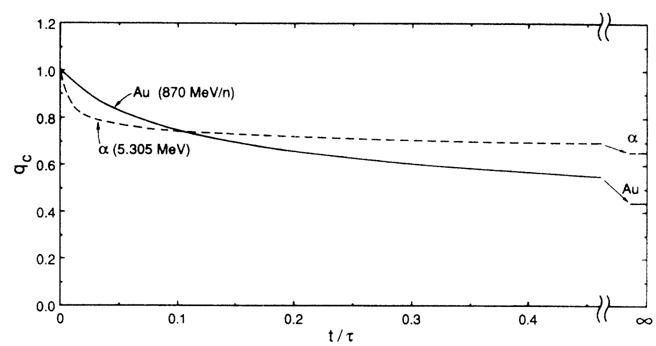

FIG. 4. The calculated evolution of quenching in the track core produced in liquid Argon by 5.305-MeV α particles (— — —) and 870-MeV/n Au ion (——). The ratio of the number of self-trapped excitons with and without quenching as a function of time is plotted. The initial radii are $a_0 = 3.9$ and 58 Å for α particles and Au ions, respectively. A specific reaction rate $k = k_{HS}/4 = 5\times 10^{-8}$ cm^3/s, $T_c/T = 0.722$ and 0.52 for α particles and Au ions, respectively, are used in the computation. The time is in units of the free exciton lifetime τ. The calculation was carried until $t = 4\tau$.

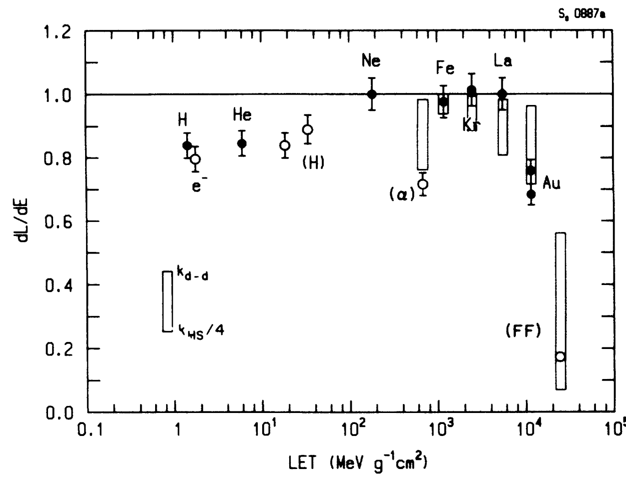

FIG. 5. The relative scintillation yield per unit energy deposition as a function of LET. Circles show experimental results[6] with error bars. Filled circles are for relativistic ions. Present calculations are shown as vertical boxes with the upper end given by a dipole-dipole collision cross section and the lower end obtained from the hard-sphere collision cross section divided by 4.

The values of q calculated by using Eqs. (2) and (13) together with the observed relative scintillation yields dL/dE are shown in Fig. 5 as a function of LET. The circles with error bars are experimental results.[6] Ions of Ne, Fe, Kr, and La do not show any significant quenching and their dL/dE values are normalized to unity. Slightly smaller values for low-LET particles such as 1-MeV electrons, 1.04-GeV/n protons, and He ions are due to escaping electrons.[6] Considerable quenching is seen for high-LET α particles, fission fragments, and Au ions. Although there is substantial qualitative agreement between experimental data and model calculations the quantitative agreement in Fig. 5 is not exact. The main reason for this is that the quenching cross section σ is not well known and we are only able to give upper and lower limits, denoted, respectively, by $\sigma_{\mathrm{HS}}/4$ and $\sigma_{d\text{-}d}$. The corresponding calculated results are contained within vertical boxes in Fig. 5. On the other hand, it should be noted that there is no other theory at present which gives a better or comparable agreement with experiment for liquid Ar. In this paper we have attempted to combine the various elements of energy deposition, diffusion, self-trapping, and biexcitonic interaction to develop a useful model for the LET dependence of specific luminescence in liquid Ar. It shows the general agreement of the track theory with a biexcitonic quenching mechanism, but that the quenching rate constant is not well known. The important finding is that the experimental results at all LET are bracketed between the limits of theoretical quenching cross sections.

As we have stated in the previous paragraph the main uncertainty in the theoretical calculation is attributable to the quenching cross section σ. It is clear that a cross section $\sim$ a few tens of Å^2 is quite reasonable to explain the order of magnitude of the quenching. As expected quenching computed with $\sigma=\sigma_{d\text{-}d}$ is too small; that computed with $\sigma=\sigma_{\mathrm{HS}}/4$ agrees well with experiments for α particles and Au ions. However, the corresponding computed value of q for fission fragments is too low, which may be attributable to δ rays penetrating the core. In our calculations the δ rays of the fission fragments were not treated separately from the core. If so done the calculated q value should be larger and nearer the experiment. For 18–40 MeV protons our calculation shows insignificant quenching, a typical value being $q=0.99$. The relative experimental value of $dL/dE=0.8$–0.9 reported for protons in this energy interval may then be attributable to escaping electrons.

Another source of uncertainty in the theoretical calculation originates from the lack of precise information about the "free" exciton lifetime τ in liquid Ar. It can be longer than 1 ps,[24,30] perhaps as large as ~ 10 ps since the luminescence of "free" excitons has been observed.[24] If we assume an exciton self-trapping time $\tau=1$ ps and use a recombination time $\tau_{\mathrm{rec}}=0.4$ ps for α particles (see Sec. II) then the α core, initially ~ 4 Å in radius, will diffuse out within the recombination time scale resulting in little quenching. As seen from Fig. 4 about 85% of the quenching is over at $t=0.4\tau$. On the other hand, for Au ions with ~ 60 Å initial core radius, considerable quenching can be expected if a slightly larger cross section is assumed. While the free exciton lifetime τ against self-trapping is not known precisely in LAr we should note that the return time for the electron can be shorter if the electron recombination occurs in a higher n-exciton state which has a larger radius. Therefore it is not unreasonable to expect that $\tau_{\mathrm{rec}} \ll \tau$, which is the basis of our model calculation.

The scintillation yield as a function of LET has been studied for many years[31] and a number of models have been proposed for quenching. Luntz[32] assumed that quenching occurs when the deposited energy density exceeds a critical value and applied the model to NaI(Tl). This model is probably inapplicable to liquid Ar because a critical value which gives some quenching for Au ions demands complete quenching for α-particle cores as seen from Fig. 3, but that is not the case.

The ion-explosion theory is also generally inapplicable to liquid Ar. It has been observed that an external electric field as low as ~ 1 kV/cm can influence the quenching of α cores in liquid Ar.[33] The field corresponds to a difference in potential energy $\sim 10^{-4}$ eV for atomic dimensions which is far weaker than that required by the ion-explosion theory. Nevertheless this fact does not completely rule out the possibility that the field-independent part of quenching is caused by ion explosion.

Salamon and Ahlen[23] introduced the diffusion of excitons and a second-order annihilation process for quenching in NaI(Tl). They assumed that the quenching is a rather slow process. The evolution of quenching was carried out until $t=10^{-8}$ s, which corresponds to the lifetime of the "trapped" exciton (or electron-hole pair) in NaI(Tl). However, as has been discussed above, quenching is a very rapid process in liquid Ar as well as in inorganic crystals.[34]

The influence of secondary electrons was not taken into account when the recombination time τ_{rec} was calculated in Sec. II. It was sufficient to show that τ_{rec} in the high-LET track cores can be much shorter than the electron thermalization time and less than 1 ps. It should be included in a refined theory. The recombination process in a dense plasma needs much more detailed discussion. We assumed that "free" excitons are responsible for quenching. Other highly excited or ionized species with a high velocity and a reasonable collision cross section are also possible candidates for quenching. Studies with very short time resolutions are needed to identify such processes.

IV. CONCLUSIONS

(i) Dependence of relative luminescence yield in Lar on particle LET and quality can be explained by a biexcitonic quenching mechanism using a track model. Loss of luminescence at low LET may be attributable to escaping electrons. (ii) On heavy-ion tracks most recombinations may be over before the self-trapping time scale. (iii) At the same LET different quenching can result for ions having different deposited energy density in the core.

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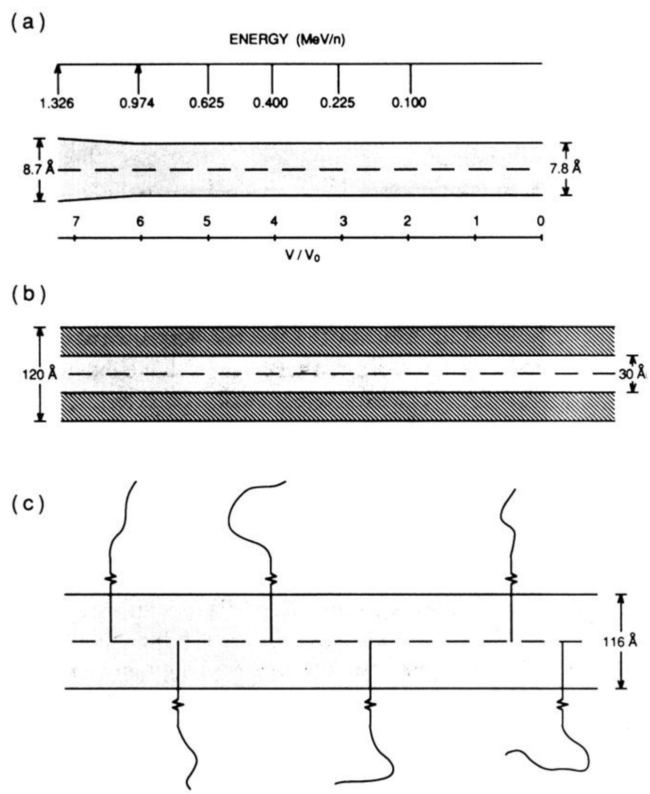

FIG. 1. The core and penumbra structure of heavy-ion tracks in LAr: (a) α particles; (b) fission fragments; (c) Au ion (870 MeV/n). The core is shown as dotted. For α particles the penumbra is a disjointed collection of emergent δ rays (not shown). These are $\sim 200-2000$ eV in energy, ejected nearly perpendicular to the track axis and have LET's somewhat less than that of the main particle. In the case of fission fragments the δ rays coalesce to form a quasicontinuous penumbra although the energy density is less than that in the core. The average δ ray on the Au ion track has energy ~ 2 keV; these are ejected nearly perpendicular to the track axis with negligible stopping or scattering within the core. The mean ranges of these δ rays are ~ 2000 Å, their LET's are orders of magnitude smaller than the main particle and they overlap little. The wavy lines indicate breaks in the path of δ rays. See text for details.