COMITATO NAZIONALE PER L'ENERGIA NUCLEARE
Laboratori Nazionali di Frascati

LNF - 70/53
10 Dicembre 1970

M. Ladu and M. Roccella: RESPONSE OF IMPURITY ACTIVATED ALKALI HALIDES SCINTILLATORS TO IONIZING PARTICLES. -

Servizio Documentazione
dei Laboratori Nazionali di Frascati del CNEN
Casella Postale 70 - Frascati (Roma)

Laboratori Nazionali di Frascati del CNEN
Servizio Documentazione

LNF-70/53

Nota Interna: n° 496
10 Dicembre 1970

M. Ladu(x) and M. Roccella: RESPONSE OF IMPURITY ACTIVATED ALKALI HALIDES SCINTILLATORS TO IONIZING PARTICLES. -

ABSTRACT. -

In a recent theory(1) the response of NaI(Tl) to energetic heavy ions ($Z \geq 5$, Energy $1 \div 10$ MeV/a.m.u.) has been calculated in good agreement with the experimental data, but a noticeable disagreement is observed for light ions like α-particle. In the present work a model is proposed to interpretate the response of impurity activated alkali-halides that follows quite closely the foreign one, where the mechanism of energy transfer by means of exciton diffusion is taken into account. If the mean exciton free path λ is $\simeq 10^{-6}$ cm at impurity concentration of about 0.1%, a satisfactory interpretation is given of NaI(Tl) response to α-particles, protons and deuterons as well as to heavier ions, and it is shown that the response to the latter is quite unaffected by exciton diffusion.

Finally characteristic dose D_o, of the foreign theory, is in our model expressed in terms of λ and this allows to give a law of dependence of luminescence efficiency to γ-rays on impurity concentration that is in agreement with experience.

(x) - Istituto di Fisica dell'Università di Cagliari.

I. - INTRODUCTION. -

The idea of saturation effect of the luminescence centers, in impurity activated alkali-halides scintillators, to interpretate decreasing of luminescence efficiency dL/dT at high specific energy loss dT/dr, was advanced by Meyer and Murray[2, 3] against the hypothesis of ionization quenching assumed in analogy with Birks theory of organic scintillators. These authors make the assumption that energy transfer occurs, mainly by radial exciton diffusion from the ion path, exciton production being uniquely related to dT/dr. In this way they find dL/dT to be a single value function of dT/dr irrespective of the nature of ionizing particle.

Data taken from experimental results of Steigert and Newman[4] on heavy ions have shown instead a dependence of dL/dT on particle nature at fixed dT/dr values and this fact was interpreted[5] in a qualitative manner in terms of energetic δ-rays escaping the saturation column of primary ion.

Recently, in analogy with theory of R. B. E. for dry enzymes and virus by Butts and Katz[6] it has been proposed that a number of phenomena produced by heavy ions must be related to the microscopic dose distribution produced by δ-rays around the ion track. This distribution depends indeed on particle nature[7]. On this assumption it was reconsidered the interpretation of formation of track in dielectric[8], the track width in nuclear emulsion[9], as well as the NaI(Tl) response to energetic heavy ions[1] obtaining a good support from the experimental data.

In particular for NaI(Tl), dL/dT has been related to the dose pattern around the track, this pattern being calculated assuming that energy lost by the ion is continuously distributed in space surrounding the track by secondary δ-rays[7]. Deviation from the linearity of the light versus ion energy at high dT/dr is then explained by saturation of the luminescence centers following an eccess of absorbed dose near the center. In this way a good agreement is found with experimental data for energetic heavy ions ($Z > 5$ energy $1 \div 10$ MeV/a. m. u.).

For α-particle a discrepancy between theory and the experimental data of order of 50% or more was observed, and the authors interpretate this discrepancy as a result from neglecting the dose inhomogeneity into volumes of the order of the sensible volumes around Tl atoms. They assimilate this assumption to the point target approximation used in the theory of R. B. E.[6] where a target of finite size was taken as puntual to calculate the activation probability from the known distribution dose function.

Although it seems quite equivalent, in the case of impurity activated alkali-halides, we prefer to say that the target, (the luminescence center), is indeed punctual but can feel dose inhomogeneity because of the exciton migration in the lattice. This migration carries to the luminescence center an excitation energy which was produced at distances of the order of the exciton mean free path from the center.

II. - THEORY. -

In principle our theory follows quite closely the way outlined by Katz and Kobetich[6] that we riassume in short.

When the dose distribution produced by secondary δ-rays can be considered uniform within the sensible volume around the centers it is assumed that the poissonian probability p(t), in a single type process, for the mission of a photon by luminescence center at distance t from the track is given by:

$$p(t) = q(1-\exp(-D(t)/D_o)) \tag{1}$$

where D(t) is the absorbed dose at distance t from the track D_o the characteristic dose which gives a probability of 63% of the maximum photon emission probability and where q is the absolute quantum efficiency for the luminescence centers.

By means of (1) if N is the number of excited centers for unitary ion path the specific fluorescence $dL/dr = qN$ is given by:

$$dL/dr = qN = qN_o \int_o^\infty 2\pi t\, dt(1-\exp(-D(t)/D_o) \tag{2}$$

where N_o is the number of luminescence centers for unitary volume i.e. the number of impurity atoms in unitary volume. The total photon emission following the passage through the lattice of a ion of kinetic energy T_o is then:

$$L = qN_o \int_o^{T_o} \sigma\, (dT/dr)^{-1}\, dT \tag{3}$$

where

$$\sigma = 2\pi \int_o^\infty t\, dt(1-\exp(-D(t)/D_o) \tag{4}$$

is the cross section for the process of excitation a luminescence center. The specific luminescence dL/dT is hence given by

$$(5) \qquad \frac{dL}{dT} = qN_o \sigma \left(\frac{dT}{dr}\right)^{-1} = \frac{qN_o}{(dT/dr)} \int_o^\infty 2\pi t\, dt(1-\exp(-D(t)/D_o)).$$

If the hypothesis of dose uniformity within volumes of linear dimension of exciton mean free path is not verified we need a different approach to the problem.

Because of impurity concentration in the scintillator, is, in all practical cases, very small, it seems reasonable to assume that direct excitation (due either to primary particle or to δ-rays) is negligible in respect to indirect excitation (by means of radiation-less energy transfer to the center from the lattice).

Then we are principally concerned with the number $\phi(t)$ of excitons crossing unitary surface, at distance t from the ion track, after the particle has passed through.

Since the transit time of the ion, and all the time constants involved in creation and migration of excitons are negligible compared with decay time of an excited luminescence center, we can suppose all the excitons reach a lattice point at the same instant so that only one of them is responsible of the excitation of the center situated in that point.

If we calls the excitation cross section of the center, the probability p(t) for such an event, when the number crossing the unitary surface is $\phi(t)$, is:

$$(1') \qquad p(t) = q(1-\exp(-S\phi(t))$$

and (2), (3), (4) and (5) becomes respectively

$$(2') \qquad \frac{dL}{dr} = qN_o \int_o^\infty 2\pi t\, dt(1-\exp(-S\phi(t)))$$

$$(3') \qquad L = qN_o \int_o^{T_o} \sigma'(dT/dr)^{-1}\, dT$$

$$(4') \qquad \sigma' = 2 \int_0^\infty t\, dt(1-\exp(-S\,\phi(t)))$$

$$(5') \qquad \frac{dL}{dT} = qN_o\sigma'\left(\frac{dT}{dr}\right)^{-1} = \frac{qN_o}{(dT/dr)} \int_0^\infty 2\pi t\, dt(1-\exp(-S\,\phi(t)))$$

To find the value of $\phi(t)$ after ionizing particle has passed, two problem must be solved:

i) How is the number of exciton produced related to the absorbed energy.

ii) How the excitons propagate through the lattice.

According with Meyer and Murray[2] we can answer to (i) as follows. Let n_e be the number of hole-electron pairs created by a charged particle. In a crystal slice we have $n_e=k(dT/dr)$. If n_o is the number of excitons resulting from electron hole recombination the recombination probability n_o/n_e is given by

$$(6) \qquad \frac{n_o}{n_e} = \frac{\alpha n_e}{1+\alpha n_e} = \frac{\alpha k(dT/dr)}{1+\alpha k(dT/dr)} .$$

Meyer and Murray[3] showed, by comparison with experimental data of dL/dT for electrons, that values of (αk) is in NaI of about $2(KeV\,cm^2/mg)^{-1}$. This allows us to put $n_o/n_e=1$, except at very low dT/dr. This means that every electron-hole pair will recombine to form an exciton. In these cases if D(t) if the absorbed dose and $n_o(t)$ the number of excitons produced in an unitary volume at a distance t from the track we have $n_o(t)=QD(t)$, Q being a proportionality constant, in all the cases we can write:

$$(7) \qquad n_o(t) = QD(t)\,\frac{(\alpha k)\,dT/dr}{1+(\alpha k)\,dT/dr)}$$

To answer question (ii) Murray and Meyer[3] solved a diffusion time-dependent equation to take into account of the depletion of unoccupied centers, Indeed in their theory the excited centers begin transparent to excitons and this leads to a dependence of the exciton mean free path on the number of excited centers. One of the results of this assumption is a marked dependence of the shape of the dL/dT vs. dT/dr curve on impurity concentration, in contrast with some experimental results

on CsI(Tl)[10, 11]. This circumstance was advanced against the model based on saturation of luminescence centers to support the model based on ionization quenching. In our model the exciton mean free path is assumed to be independent on the number of excited centers. We will show qualitatively in the next, that in this way the dependence of the shape of dL/dT vs. dT/dr curve can be considerably attenuated.

The law of exciton migration assumed in the next, is

$$p(r) = \frac{e^{-r/\lambda}}{4\pi r^2} \tag{8}$$

where λ is the exciton mean free path and where p(r) is the probability that an exciton produced at the origin cross the unit surface at a distance r.

The law (8) for exciton migration has been introduced by Birks to explain the surface quenching effect[11, 12] in organic scintillator, i.e. the dependence of dL/dT on distance from scintillator surface near their free surface.

In organic scintillators the value of λ can be deduced by means of this effect and values of some microns have been found. Since the values of λ was can expect for impurity activated alkali-halides at the usual values of the impurity concentrations ranges from some tens to one or two hundreds of angtrons, the surface quenching should be negligible in these cases. Hence we will deduce the value of λ from the best fit using the experimental data of the response of NaI(Tl) to α-particle, protons and deuterons.

In the case of protons up to $10 \div 20$ MeV and heavier ions, with the aid of (7) and (8) the expression for $\phi(t)$ using polar coordinates (r, θ, φ) having origin in the point under consideration at distance t is:

$$\phi(t) = \int dV\, Q\, D(t')\, e^{-r/\lambda} / 4\pi r^2 \tag{9}$$

where D(t') is the absorbed dose at distance $t' = (t^2 + r^2 \sin^2\theta + 2rt\sin\theta)^{1/2}$ from the ion track and where the integration is extended to the entire scintillator i.e. practically to infinity.

For values of λ very small the integral (9) can be performed taking D(t) constant. In this case we have:

$$\phi(t) = \lambda\, Q\, D(t)$$

and (1')÷(5') become identical to (1)÷(5) respectively if we put:

$$D_o = (Q \lambda S)^{-1} \tag{10}$$

The (10) that relate D_o to λ, us we shall see leads to interpret in a direct and very simple way the dependence of luminescence efficiency to γ-rays on impurity concentration.

In the next section III we shall consider separately the problem of removing point target approximation for heavy ions and light ions ($Z < 5$).

III. - NaI(Tl) RESPONSE TO VARIOUS IONS. -

III. a. - Response to heavy ions. -

To remove point target approximation for heavy ions we try to obtain only an approximate expression for σ as long as we must think that the error introduced by this approximation does not excede, in these cases, some per cent.

If D(t') shows only a smooth dependence on r with respect to $e^{-r/\lambda}$ (and this is the case when $t > \lambda$) we have

$$\phi(t) = Q\lambda\left[D(t) + \frac{\lambda^2}{3}\left(\frac{1}{t}\frac{dD}{dt} + \frac{d^2D}{dt^2}\right) + \theta(\lambda^4)\right] \tag{11}$$

Now since $\phi(t)$ occurs in the expression of σ' only in the exponential, factor when $D(\lambda) \geq 2 \div 3\ E_o$ we can introduce (11) in (4') without making any relevant error in σ, irrespective, up to large extent, of the error that effect ϕ when $t < \lambda$.

In performing integration (4') we will use for D(t) the approximate expression;

$$D(t) = \frac{A}{t}\left(\frac{1}{t} - \frac{1}{T}\right) \tag{12}$$

used in ref. (6) A and T being constant depending on the value of $\beta = v/c$. Then from (11) and (12) we obtain

$$\sigma' \cong \sigma + \frac{4}{3}\pi\lambda^2 - \frac{1}{3}\pi\lambda^2\left[\frac{\lambda}{T}\left(\frac{D(\lambda)}{D_o}\right)^{1/2} - \frac{\lambda^2}{T^2}\frac{D(\lambda)}{D_o}\right] \tag{13}$$

where σ is given by (4) and $D(\lambda)$ is the value assumed by D(t) at a distance λ from the track.

If we replace in (10/values of λ between $(0.5 \div 1)10^{-6}$ cm, we can see that σ' differs from σ only by some per cent in the energy range we are concerned with, when $Z > 5$.

In Fig. 1 we reported σ' versus MeV/a.m.u. (full lines) for $Z = 6$ and $Z = 8$ and $\lambda = 10^{-6}$ cm, together with σ (dotted lines) calculated by Katz and Kobetich[1] for the same values of Z.

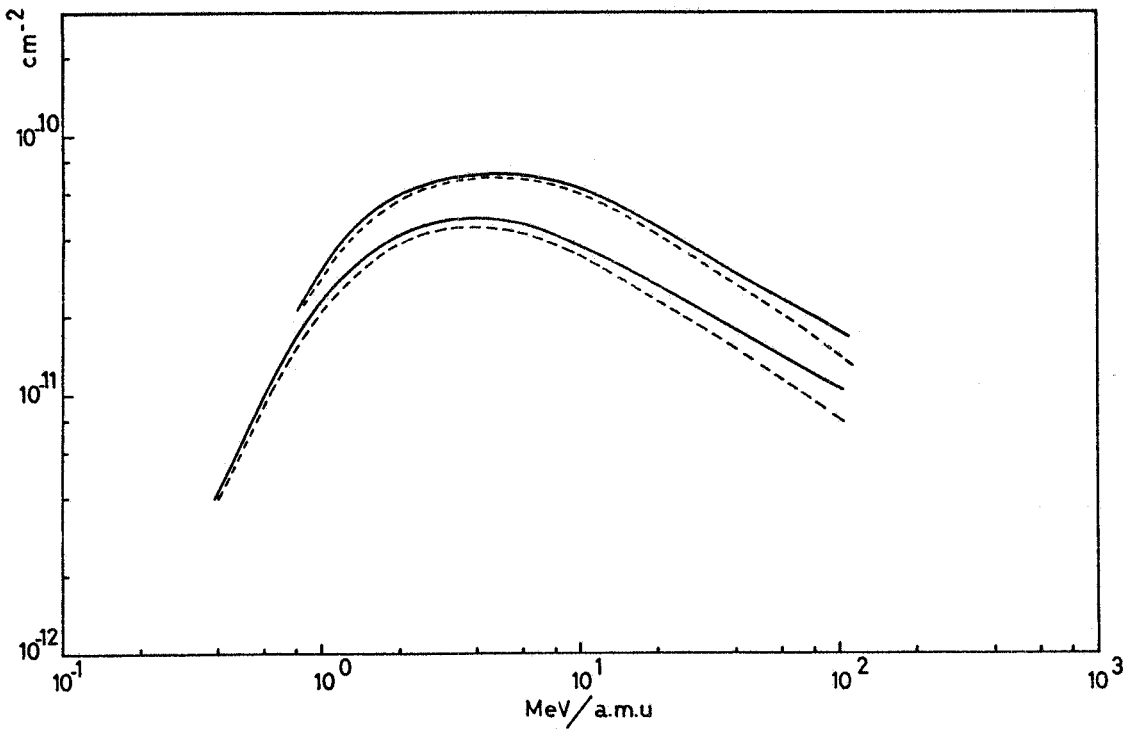

FIG. 1 - Excitation cross sections for Z = 8 (upper curves) and Z = 6 (Lower curves). By Katz and Kobetich (dotted lines) and present work (full lines).

III.b. - Response to light ions. -

Since for α-particles, protons and deuterons, the conditions under which (13) has been obtained are not quite well verified, except at very Low values of β, for these particles we need to perform numerical integration of (9).

λ range was from 0.5 to 1×10^{-6} cm. The dose distribution D(t) around the track has been obtained assuming that all the energy lost by ionizing particle is distributed around the track by secondary δ-rays following the pattern calculated by Katz and Kobetich[1].

D(t) can be obtained from the relation

(14) $$D(t) = z^2 d_1(t) + z^3 d_2(t)$$

where z is the effective charge number related to Z by

$$z = Z\left[1-\exp(-125/\beta Z^{-2/3})\right]$$

$d_1(t)$ and $d_2(t)$ being obtained from ref. (1). In analogy with (14) we put

(15) $$\psi(t) = S\phi(t) = z^2 \psi_1(t) + z^2 \psi_2(t)$$

where $\psi_1(t)$ and $\psi_2(t)$ are given by

$$\psi_1(t) = \frac{1}{4\pi D_o}\int dV \frac{d_1(t')\, e^{-r/\lambda}}{r^2}; \qquad \psi_2(t) = \frac{1}{4\pi D_o}\int dV \frac{d_2(t')\, e^{-r/\lambda}}{r^2}$$

As for $z > 5$ the first term of (12) predominates widely we have computed $\psi_1(t)$ only. In Fig. 2, $\psi_1(t)$ is reported as a function of t for $\lambda = 10^{-6}$ cm and in Fig. 3 curves of σ' versus MeV/a. m. u. calculated by (4') using data of Fig. 2 are reported for α-particles, protons and deuterons. The curve calculated by Katz and Kobetich for α-particles is also reported in Fig. 3 in order to make a direct comparison with the value we have obtained.

Finally in Fig. 4 theoretical response curve are given together with experimental data taken from Eby and Jentshke[13] for the optimal value of the impurity concentration. In the same figure the dotted curve represents that one obtained by R. Katz and J. Kobetich for α-particles.

IV. - DEPENDENCE OF ABSOLUTE SCINTILLATION EFFICIENCY TO γ-RAYS ON IMPURITY CONCENTRATION. -

We now turn to the problem of obtaining dL/dT relative to γ-rays, versus impurity concentration. If we are concerned with a ionizing particle i we have

(17) $$\frac{dL_i}{dT} = q[Y]\frac{\sigma_i}{dT/dr}$$

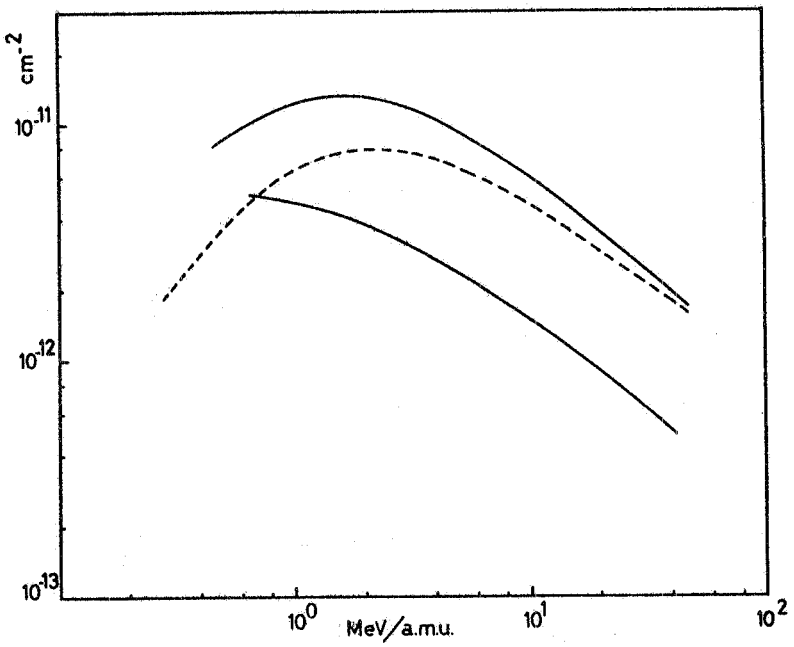

FIG. 2 - ψ_1 versus the distance t from the ion track at the selected value of $\lambda = 1 \times 10^{-6}$ cm.

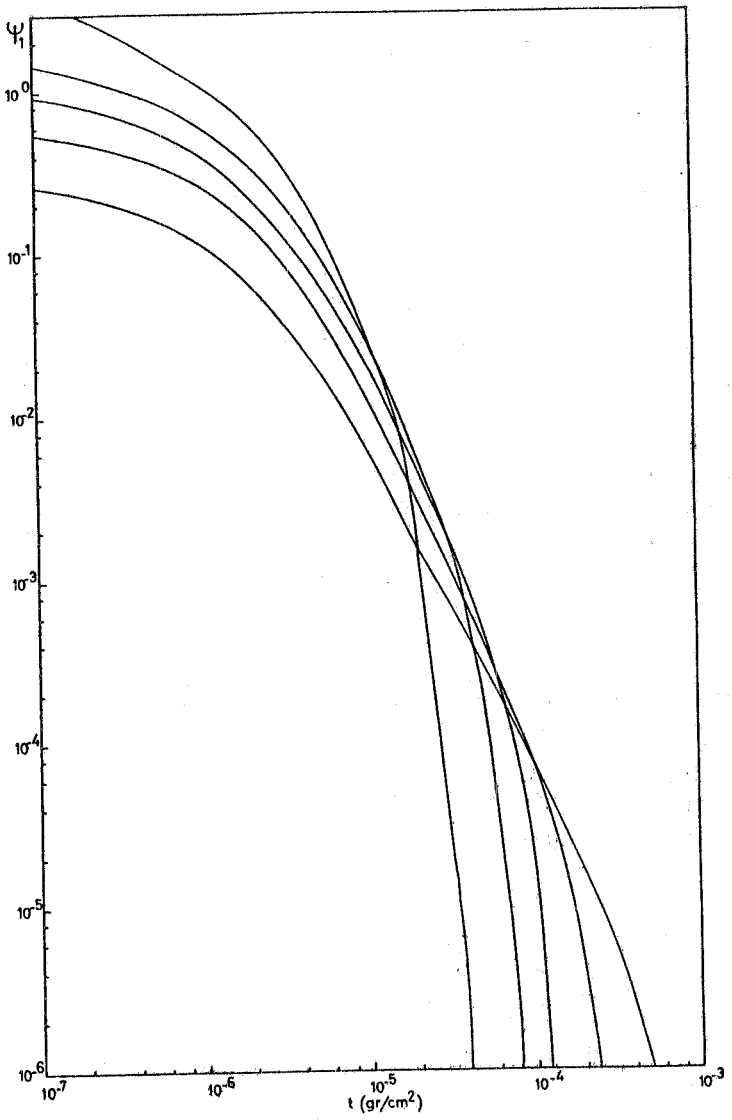

FIG. 3 - Excitation cross sections for α particles (upper curve) protons end deuterons (lower curve). Dotted line represents the curve calculated by Katz and Kobetich for α-particles.

where σ_i is obtained by (4') using the dose pattern relative to particle i, $[Y]$ being the impurity concentration in per cent.

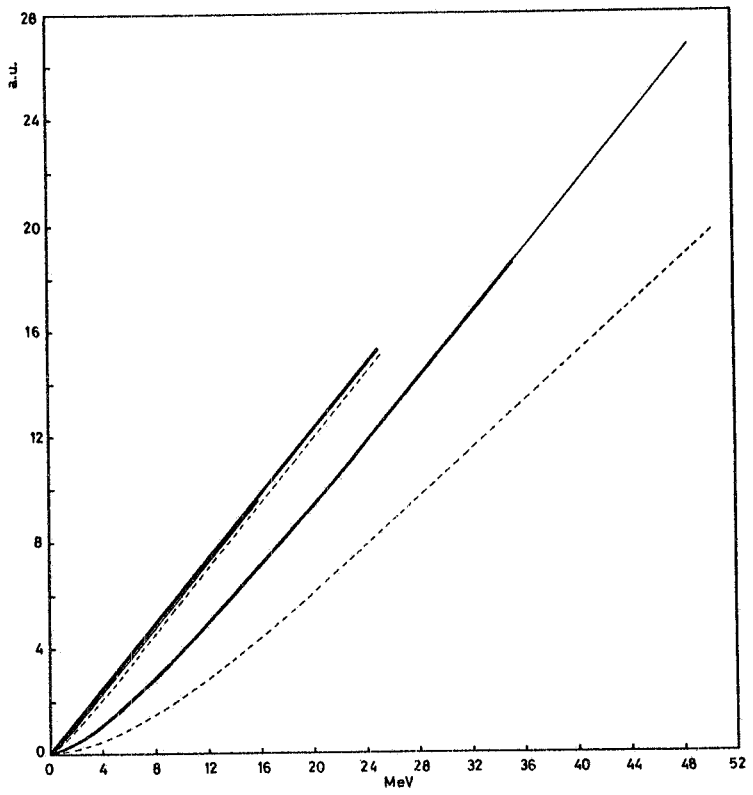

FIG. 4 - Response of NaI(Tl) versus energy. Heavy lines; experimental response[4] to protons and deute rons (upper curve) to α - -particles lower curve. Light lines: response to protons present work (upper line), response to α- -particle present work (lo wer curve). Dotted lines: response to deuterons pre sent work (upper curve). Response to α-particles Katz and Kobetich (lower- -curve).

In considering the response to γ-rays we can assume an uniformly distributed dose except at very high exposure rates. (Indeed in these cases we ever have local closes $D \ll D_o$ during exposure times of order or less of the luminescence decay time). With this assumption, using (7) and (10), eq. (17) becomes

$$(18) \qquad \frac{dL_i}{dT} = \frac{q[Y]}{D_o(dT/dr)} \int_o^\infty D(t)\, 2\, t\, dt = \frac{q[Y]}{D_o} = q[Y] Q\, S \frac{(\alpha k) dT/dr}{1+(\alpha k) dT/dr}$$

being $\int_o^\infty D(t)\, 2\pi t\, dt = dT/dr$. The constant q which is the probability for photoemission from an excited luminescence center will be expressed according with Birks[10] by

$$(19) \qquad q = \frac{q_o}{1 + S_{cy}[Y]}$$

where S_{cy} describes competition between processes of concentration quenching, luminescence, and internal quenching and where q_o is the luminescence efficiency of solute extrapolated to zero concentration of impurity. The exciton mean free path λ shall be expressed by $\lambda = (S_x[X]+S_y[Y])^{-1}$ where S_x and S_y represent ecciton capture cross section by solvent X (the lattice sites) and solute Y (the impurity).

If $[X]$ and $[Y]$ are solvent and solute concentration in per cent. Because $[Y] \ll 1$ in all pratical cases, we have $[X] = 1 - [Y] \simeq 1$ and:

$$\lambda = \frac{1}{S_x + S_y[Y]} \tag{20}$$

By (18, 19, 20) we have at low dT/dr

$$\frac{dL}{dT} \propto \frac{\alpha k\, dT/dr}{1+\alpha k\, dT/dr} \cdot \frac{[Y]}{1+S_{yx}[Y]} \cdot \frac{1}{1+S_{cy}[Y]} \tag{21}$$

Where $S_{yx}=S_y/S_x$. It has been shown[11] that although a formula like (21) has been obtained only for organic binary solution eq. (21) is also in good agreement with experience relative to NaI(Tl)[14] if are taken $S_{yx} = 8 \times 10^3$ and $S_{cy} = 60$. Moreover at $[Y] \ll 1$ the theoretical formulae obtained for impurity activated alkali halides by Johnson and Williams[15, 16] can be easily reduced to (21).

V. - CONCLUSION. -

In Fig. 5 dL/dT obtained from (5') and (7) is given as a function of dT/dr (full lines) up to 500 KeV cm^2/mg and is compared with experimental data (full point), Murray and Meyer theory (point-dotted line), and Katz and Kobetich theory for α-particles (dotted line). As we can see there is a good agreement with Murray and Meyer result in this range of dT/dr and both agree quite well with experimental result.

Perhaps a better agreement is obtained in our theory because of sensible splitting we obtain of dL/dT versus dT/dL for protons and α-particle. Note instead the noticeable discrepance between Katz-Kobetich result for α-particle and experimental data.

At higher value of dT/dr the dependence of dL/dT on particle nature begin more and more marked. For these higly ionizing particles Katz and Kobetich theory agree very well with experience[1] and, as our theory begin quite insensitive to exciton diffusion (see (13) and Fig. 1), our results become similar to those obtained by Katz and Ko

betich. In conclusion we can say that the values of dL/dT found by our model agree well with experience in a wide range od dT/dr and for very different particles like electrons and heavy ions.

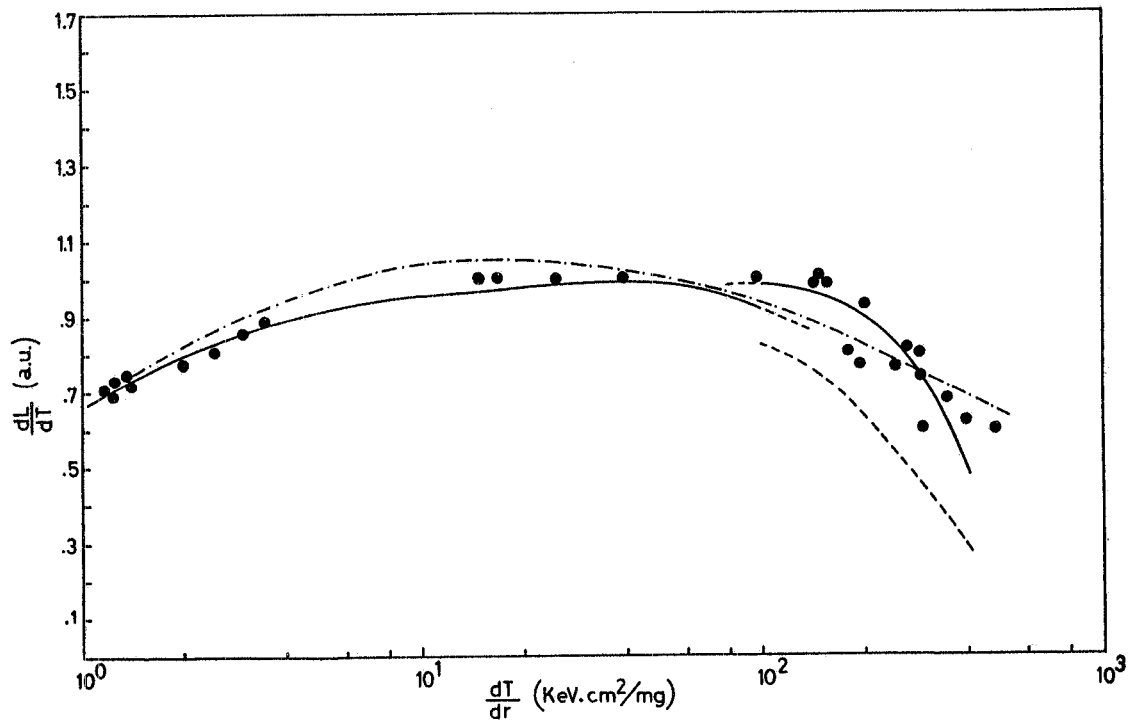

FIG. 5 - Luminescence efficiency of NaI(Tl) versus specific energy loss. Experimental results[10] fulf points, Mayer and Murray model point-dotted line, present work (full line), Katz and Kobetich (dotted line).

At the end we turn out to the question of the dependence of the shape of dL/dT versus dT/dr on [Y] concentration or in an equivalent way of the dependence of the shape of dL/dT versus [Y] concentration on dT/dr. If we look now at eq. (21) we can see that with the hypothesis of depletion of unoccupied centers made by Meyer and Murray all occurs as if the effective [Y] in the expression of eq. (21) would decrease with increasing dT/dr leading to a dependence of the shape of dL/dT versus [Y] on dT/dr.

As long as in our model λ is taken to be independent on concentration of excited centers and as a consequence, on dT/dr, we can infer that this dependence in our model would be smoothed out with to respect Murray and Meyer model.

REFERENCES. -

(1) - R. Katz and E. J. Kobetich, Phys. Rev. 170, 397 (1968).
(2) - A. Meyer and R. B. Murray, IRE Trans. Nuclear Sci. 7, n. 2-3, 22 (1960).
(3) - R. B. Murray and A. Meyer, Phys. Rev. 122, 815 (1961).
(4) - E. Newman and F. E. Steigert, Phys. Rev. 118, 1575 (1960).
(5) - R. B. Murray and A. Meyer, IRE Trans. Nuclear Sci. 9, n. 3, 33 (1962).
(6) - J. Butts and R. Katz, Radiation Res. 30, 855 (1967).
(7) - R. Katz and E. J. Kobetich, Phys. Rev. 170, 391 (1968).
(8) - R. Katz and E. J. Kobetich, Phys. Rev. 170, 401 (1968).
(9) - R. Katz and E. J. Kobetich, Phys. Rev. 170, 405 (1968).
(10) - S. B. Birks, Theory and Practice of scintillation counters (Pergamon Press, 1964).
(11) - R. Gwin, Oak Ridge National Laboratori, Report ORNL 3354 (1962).
(12) - J. B. Birks, Phys. Rev. 86, 569 (1952).
(13) - F. S. Eby and W. K. Jentschke, Phys. Rev. 96, 911 (1954).
(14) - J. A. Harshaw, H. C. Kremers, E. C. Stewart, E. K. Warburton and S. O. Hay, U. S. Atomic Energy Commission, Report NYO-1577 (quoted by Murray and Meyer 1961) (1952).
(15) - P. D. Johnson and F. E. Williams, J. Chem. Phys. 18, 323 (1950 a).
(16) - P. D. Johnson and F. E. Williams, J. Chem. Phys. 18, 1477 (1950 b).