June 1968

Response of NaI(T1) to Energetic Heavy Ions

Robert Katz
University of Nebraska-Lincoln, rkatz2@unl.edu

E. J. Kobetich
University of Nebraska-Lincoln

Response of NaI(Tl) to Energetic Heavy Ions*

ROBERT KATZ AND E. J. KOBETICH
Behlen Laboratory of Physics, University of Nebraska, Lincoln, Nebraska

(Received 18 December 1967)

Experimental values of the relative heights of scintillation pulses generated in NaI(Tl) crystals, by heavy ions ($Z>5$) of energy 1–10 MeV/amu, agree well with computed relative cross sections for photon production, from a theory based on the assumption of a one-hit response to the spatially distributed dose of ionization energy, and a characteristic dose of 4×10^7 erg/cm^3 for this material. Discrepancies between theory and experiment for He bombardments arise from the theoretical neglect of the nonlinear dose variation over the sensitive volume surrounding each Tl atom. Similar discrepancies arising from the neglect of molecular volume occur in the theory of heavy-ion inactivation of dry enzymes and viruses, which forms the basis for the present work.

I. INTRODUCTION

FOR all scintillation media there exists a decreasing scintillation efficiency with increasing specific energy loss of the primary particle. In their analysis of the response of alkali-halide scintillators, Meyer and Murray[1] took the differential light output per unit energy loss, called the scintillation efficiency $d\mathfrak{L}/d\mathfrak{E}$ to be a single valued function of the specific energy loss of the incident particle $d\mathfrak{E}/d\mathfrak{x}$. The total emitted light per unit length of path of the primary particle was then taken to be composed of two contributions, one from the highly ionized "primary column" (of adjustable diameter) and the other from those δ rays escaping the primary column, each evaluated according to its specific energy loss.

The present work returns to an earlier view, that the characteristic response of scintillation media to heavy ions is due to the saturation of luminescence centers, and that the specific energy loss is not an appropriate parameter for describing these effects, for it contains no knowledge of the spatial distribution of ionization energy. The ultimate generation of photons by luminescence centers may be traced back to the deposition of energy in the medium by δ rays (all secondary electrons ejected by the passing ion). Energy transfer processes carry the deposited energy from the passive matrix to the luminescence center. Competition, between radiationless decay of excitation energy in the matrix and energy transfer, limits the energy transfer to a characteristic distance, so that a *sensitive volume* may be associated with each luminescence center. As implied by such a model, it is indeed observed that the pulse height for a particular bombardment first increases nearly linearly with luminescence center concentration, and then saturates when the volume per luminescence center approximates the sensitive volume.

The energy deposited by δ rays is assumed to pass into a number of channels, some of which excite the matrix appropriately for subsequent transfer to a luminescence center. Thus the dosage of deposited energy in elementary subvolumes about the ion's path is a suitable parameter for characterizing ultimate photon production. Since the ion passes a sensitive volume in a time interval short compared to the mean life of an excited luminescence center, no more photons are produced by many excitation events in the sensitive volume than by a single event. The distribution of excitation events is taken as random, in small subvolumes, and the response of the medium is then appropriate to a one-or-more-hit cumulative Poisson distribution.

The assumption that the excitation events are random is somewhat stronger than is required. A test for the randomness of random-number sequences, suggested by Katz,[2] makes use of the departure of the tally of n-hit events from the predictions of the binomial distribution. The tally of one-or-more-hit events in cells the size of a sensitive volume is a weak test of randomness, which may be satisfied even though the ionizing events are correlated to the tracks of δ rays.

Very similar problems arise in radiobiology. Enzyme and virus molecules are very specific in their biological functions. The rupture of a single bond inactivates the molecule. The sensitive volume is the physical volume of the molecule.

The theory of Meyer and Murray on the response of scintillation media resembles the target theory of Lea,[3] as extended by Hutchinson and Pollard,[4] and others.[5–8]

The present theory of the response of scintillation media follows closely upon the theory of relative biological effectiveness for the heavy-ion bombardment of dry enzymes and viruses of Butts and Katz.[9]

* Supported by the U.S. Atomic Energy Commission and the National Science Foundation.

[1] A. Meyer and R. B. Murray, Phys. Rev. **128**, 98 (1962).

[2] R. Katz, Bull. Am. Phys. Soc. **11**, 113 (1966).

[3] D. E. Lea, *Actions of Radiations on Living Cells* (Cambridge University Press, New York, 1962), 2nd ed.

[4] F. Hutchinson and E. Pollard, in *Mechanisms in Radiobiology*, edited by M. Errera and A. Forssberg (Academic Press Inc., New York, 1961), Vol. 1, Chap. 1, Pt. 2.

[5] G. W. Dolphin and F. Hutchinson, Radiation Res. **13**, 403 (1960).

[6] P. E. Schambra and F. Hutchinson, Radiation Res. **23**, 514 (1964).

[7] D. J. Fluke, T. Brustad, and A. C. Birge, Radiation Res. **13**, 788 (1960).

[8] D. J. Fluke and F. Forro, Jr., Radiation Res. **13**, 305 (1960).

[9] J. J. Butts and R. Katz, Radiation Res. **30**, 855 (1967).

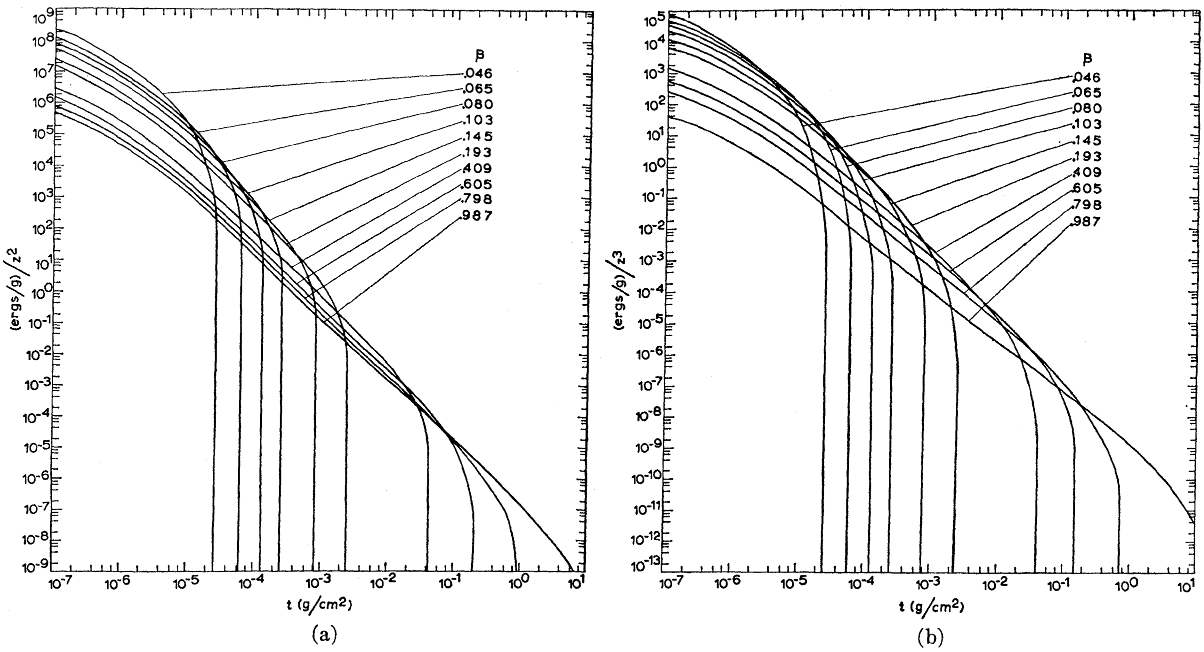

FIG. 1. Spatial distribution of ionization energy in NaI. To find the energy dosage deposited at radius t (erg/g) by an ion of effective charge z moving at speed βc, the value given in curve (b) must be multiplied by z, added to the value obtained from (a), and the sum multiplied by z^2. The effective charge number z may be obtained from the atomic number Z by the expression $z=Z[1-\exp(-125\beta Z^{-2/3})]$. See Ref. 11.

II. THEORY

According to Eby and Jentschke,[10] the pulse height generated by α particles in NaI(Tl) crystals reaches a maximum value near a Tl mole fraction of about 10^{-3}, implying that the mean spacing of Tl atoms is about 10 lattice spacings, or 65 Å. We take this to be the mean diameter of the sensitive volume. In a crystal in which the mean volume per luminescence center is greater than the sensitive volume, the cumulative one-or-more-hit Poisson distribution suggests that the probability per luminescence center for the emission of a photon from a scintillating medium which has absorbed a uniform dose of ionization energy E is given by the expression

$$P=1-\exp(-E/E_0), \tag{1}$$

where E_0 is the dose for excitation of 63% of the luminescence centers of a particular medium.

The number of interactions within a cylindrical shell of thickness dt, mean radius t, and length l (sufficiently short so that the variation in the ion's speed within the cylinder may be neglected) is given by the product of the volume of the shell, the "effective" number of scintillation centers per unit volume (compensating for overlap of sensitive volumes at high Tl concentration), and the probability for photon emission. If $\mathfrak{N}$ is the effective number of Tl sites per unit volume, the number of activated sites within the shell, $d\mathcal{N}$, is given by the expression

$$d\mathcal{N}=\mathfrak{N}\times 2\pi t l dt[1-\exp(-E/E_0)]. \tag{2}$$

The total number of activated luminescence centers $\mathcal{N}$ arising from the passage of an ion is found by integrating Eq. (2) over the volume containing all the deposited ionization energy. Dividing the number of activated centers within this volume by the number of possible sites per unit area in the distance l, the cross section for the process is

$$\sigma=\mathcal{N}/\mathfrak{N}l=2\pi\int_0^\infty [1-\exp(-E/E_0)]t dt. \tag{3}$$

Since the number of activated centers per unit length $\mathcal{N}/l$ is proportional to the emitted light per unit path length, theoretical values of σ are proportional to experimental values of $d\mathfrak{L}/d\mathfrak{x}$, and

$$\int_0^{\mathfrak{E}} \sigma(d\mathfrak{E}/d\mathfrak{x})^{-1}d\mathfrak{E}$$

is proportional to $\mathfrak{L}$.

In this development, an assumption of small targets is implicit. The entire sensitive volume is assumed to experience the same dose, and thus has the same activation probability. The calculation is then rigorously valid for point targets. When the point-target theory is applied to extended volumes, it is implied that the dose at the luminescence center equals the average dose over the sensitive volume. This is a poor approxi-

[10] F. S. Eby and W. K. Jentschke, Phys. Rev. 96, 911 (1954).

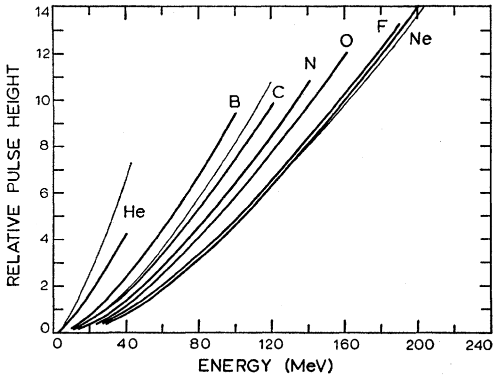

FIG. 2. Experimental values of the relative pulse height $\mathfrak{L}$ generated in NaI(Tl) by ions of incident energy $\mathfrak{E}$ from Newman and Steigert (see Ref. 12) (light lines) compared to theory (heavy lines) computed at a critical dose of 4×10^7 erg/cm³. Theory and experiment agree sufficiently closely for B, N, O, F, and over most Ne energies that the light lines are obscured by the heavy lines.

mation close to the ion's path. The point-target theory always underevaluates the contribution to the cross section in this region of the medium. Where the dose E exceeds the critical dose E_0 at a distance of about three times the radius of the sensitive volume from the ion's path, the close-in luminescence sites are saturated, and the point-target approximation yields good results, as shown by Butts and Katz.[9] The point-target theory may be expected to underestimate the cross sections for low-Z bombardments.

III. RESULTS

To apply Eq. (3) to the computation of the cross section for photon emission, it is first necessary to find the spatial distribution of ionization energy. This function has been calculated for NaI, following the

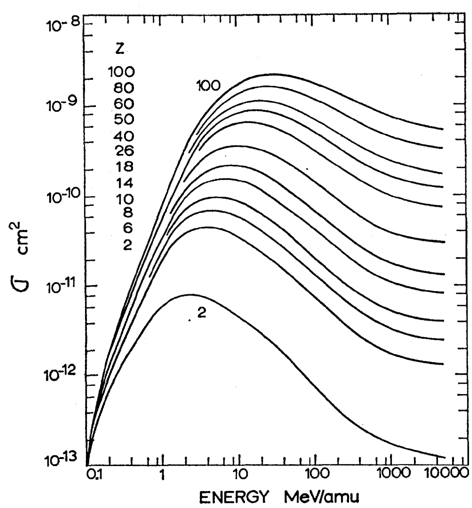

FIG. 3. Theoretical values of the cross section σ for activation of a luminescence center in NaI(Tl) versus the ion energy $\mathfrak{E}$, using the point-target approximation, valid for $\sigma\geq3\times10^{-11}$ cm².

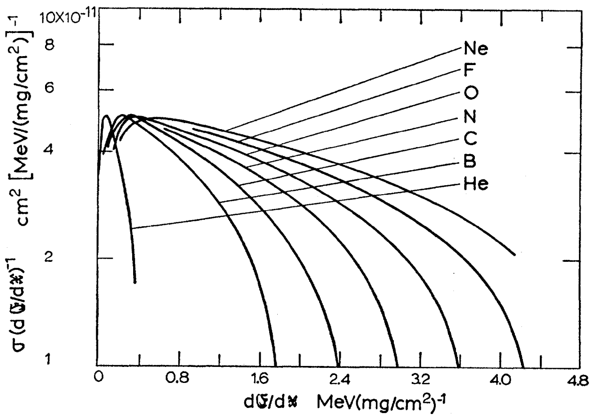

FIG. 4. Theoretical plot of $\sigma(d\mathfrak{E}/dx)^{-1}$ versus $d\mathfrak{E}/dx$.

procedure of Kobetich and Katz,[11] and is shown in Fig. 1.

In application of the one-hit theory to radiobiology, there are experimental data for both the cross sections for heavy-ion bombardment and the response of the particular molecules to a uniform dose of γ rays, from which the characteristic dose, there called the D-37 dose, can be determined.

For scintillation crystals, the characteristic dose must be found by comparing the relative pulse height predicted by the theory for a variety of characteristic doses to available experimental data. Experimental values of the relative pulse height $\mathfrak{L}$ generated in NaI(Tl) crystals by a variety of ions, from Newman and Steigert,[12] are compared to theory in Fig. 2, after

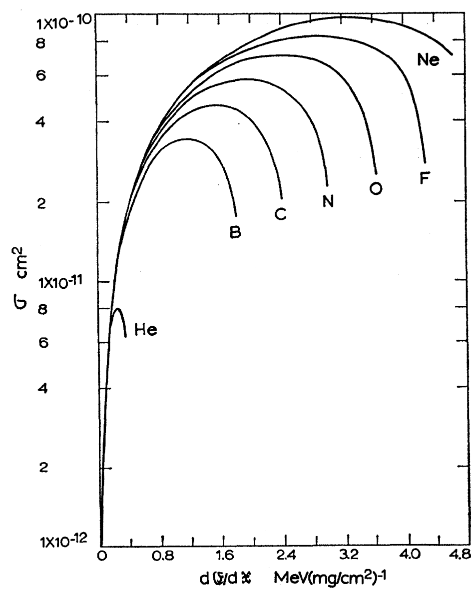

FIG. 5. Theoretical plot of σ versus $d\mathfrak{E}/dx$.

[11] E. J. Kobetich and R. Katz, preceding paper, Phys. Rev. **170**, 391 (1968).

[12] E. Newman and F. E. Steigert, Phys. Rev. **118**, 1575 (1960).

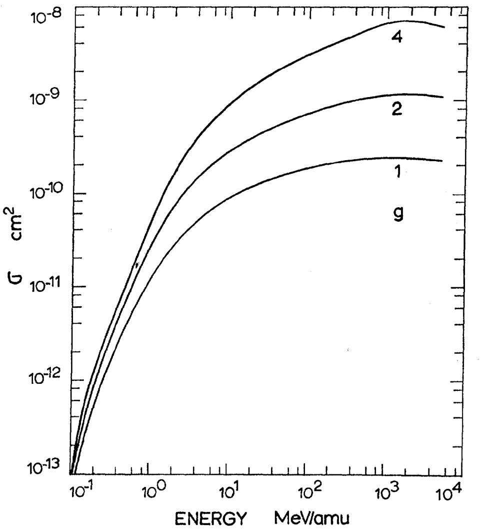

FIG. 6. Theoretical cross sections for luminescence center activation for a spectrum of magnetic monopoles of pole strength 1, 2, and 4× the strength of a Dirac monopole ($g=137e/2$).

normalization by a multiplicative factor setting theory equal to experiment for 100 MeV neon ions, with the choice of the critical dose as $E_0=4\times10^7$ erg/cm³, corresponding to about 3 eV/(sensitive volume). By comparison of theory with experiment, the critical dose can be set to about 25%.

Theoretical values of the cross section as a function of the ion energy, with the atomic number Z of the incident ion as parameter, are shown in Fig. 3. Following the practice of earlier investigators, theoretical values of $\sigma(d\mathfrak{E}/d\mathfrak{x})^{-1}$ (proportional to $d\mathfrak{L}/d\mathfrak{E}$) are plotted against $d\mathfrak{E}/d\mathfrak{x}$ in Fig. 4, while σ (proportional to $d\mathfrak{L}/d\mathfrak{x}$) is plotted against $d\mathfrak{E}/d\mathfrak{x}$ in Fig. 5. Specific energy loss data for NaI were obtained from Newman and Steigert[12] (for Fig. 2) and from Barkas and Berger,[13] as appropriate.

Experimental data for He and C disagree with theory in Fig. 2. No explanation is offered for the case of C ions, for theory and experiment agree for bombardments with ions of adjacent Z on either side of C. The disagreement between theory and experiment for He bombardment arises from the use of the point-target theory.

Our earlier discussion of the validity of the point-target approximation stressed the ratio of the dose of deposited ionization energy to the critical dose at a distance of about three sensitive volume radii from the ion's path. At this distance the critical dose is attained for ions of $Z\geqq6$, at 10 MeV/amu. From Fig. 2, theory and experiment agree well for B, at this energy, implying that this criterion is conservative. At 10 MeV/amu the dose at 100 Å is 3.7×10^6 erg/cm³ for He and is 2.3×10^7 erg/cm³ for B, as compared to the critical dose of 4×10^7 erg/cm³. Stating the validity criterion in another way, the point-target theory is satisfactory for $\sigma\geqq3\times10^{-11}$ cm², and yields an underestimate of relative pulse height below this value.

For completeness, theoretical cross sections for a spectrum of magnetic monopoles are shown in Fig. 6. Note that the cross section for a high-energy pole increases with energy, in contrast to the behavior of a high-energy ion.

IV. DISCUSSION

Though the details of the processes of matrix activation, energy transfer, and light emission vary among alkali-halide crystals, and among organic-liquid scintillators, the basic outline presented here for NaI(Tl) should remain unaltered for many of these media. Similar effects have been observed in CsI(Tl), for example, by Gwin and Murray,[14] as have been discussed here for NaI(Tl). The characteristic effects of heavy ions in scintillating media of all types can be expected to correlate with the existence of a sensitive volume, with its associated characteristic dose, and with the transverse distribution of ionization energy. Since the intensity of different emission bands varies with dose intensity, as shown for CsI(Tl) by Gwin and Murray[15] and for organic liquids by Carter, Christophoru, and Abu-Zeid,[16] a polychromatic structure must be displayed by the luminescent pulses from heavy ions which is characteristic of the ion's speed and charge.

[13] W. H. Barkas and M. J. Berger, Natl. Acad. Sci.—Natl. Res. Council, Publ. **1133**, 103 (1964).

[14] R. Gwin and R. B. Murray, Phys. Rev. **131**, 501 (1963).

[15] R. Gwin and R. B. Murray, Phys. Rev. **131**, 508 (1963).

[16] J. G. Carter, L. G. Christophoru, and M-E. M. Abu-Zeid (to be published).