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Track-Effect Theory of Scintillation Efficiency

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A theoretical account is presented of the pulse-height characteristics of NaI(Tl) scintillation counters subjected to energetic heavy-ion bombardment ($Z \geq 5$, $E/A = 1-10$ MeV/nucleon) at room temperature. The falling off of scintillation efficiency dL/dE with decreasing energy and the charge dependence at fixed energy are simultaneously accounted for by introducing the concept of a cylinder of high energy-deposit density surrounding the particle track. Inside the cylinder competitive events, favored by high ionization density, are assumed to dominate those which produce the characteristic luminescence emission. Agreement with experiment is best for high-Z particles. Cylinder radii vary over the range $110 \lesssim R_c(Z, v) \lesssim 390$ Å. Estimates of the fraction of the total energy loss available for efficient light production yield the values $0.20 \lesssim F_0(Z, v) \lesssim 0.50$, while the critical value of energy-deposit density defining the high-density cylinder is approximated to be 5.32×10^7 erg/cm^3. Also, a brief discussion is presented regarding interpretation of the heavy-ion pulse-height characteristics of pure alkali halides at low temperature, and those of anthracene and NE 102 plastic scintillators, in terms of the track-effect theory.

I. INTRODUCTION

The purpose of this paper is to provide a theoretical account of the response of activated alkali iodide scintillation counters to room-temperature bombardment by energetic heavy ions and, in so doing, to present a theory applicable to a fairly wide range of scintillating crystals. Treated explicitly are the data of Newman and Steigert[1] for NaI(Tl) corresponding to bombardment with B^{10}, C^{12}, N^{14}, O^{16}, F^{19}, and Ne^{20} ions of incident energies ranging from approximately 1 to 10 MeV/nucleon. The curves displaying relative pulse heights, shown in Fig. 1, are linear at the higher energies, and become distinctly nonlinear as E decreases. The direction of curvature implies a systematic falling off of scintillation efficiency dL/dE with the slowing down of a particle. Also apparent is a dependence of pulse height on particle identity, such that the lighter the ion, the greater is the total light output for the same total energy loss.

Detailed explanations have been offered to account for these features.[2–4] However, the treatments of Refs. 2 and 4 include explicit assumptions regarding luminescence mechanisms which have been demonstrated to be invalid or, at best, highly dubious. Also, several processes which are operative during the penetration of a highly ionizing particle—processes which are likely to have a profound effect on the luminescent response—are disregarded. The treatment of Ref. 3 suffers from difficulties of a somewhat different nature which, along with the above points, are discussed in the paper. Further examination of the problem would appear to be in order.

The present formulation incorporates the so-called "track-effect" profile of energy deposit about the path of a penetrating ion.[5] An imaginary cylinder surrounding the particle track is employed to partition the crystal into regions of high and low energy-deposit density. Associated with each region is a corresponding contribution to the total scintillation efficiency. Upon consideration of events favored by high ionization density, e.g., electron-hole recombination and radiation-damage and lattice-heating effects, and upon consideration of the competitive role of such events with respect to luminescence, the assumption is made that dL/dE receives a negligible contribution from within the high-density region. In contrast, the response to energy deposited outside the high-density cylinder is assumed to be linear. Thus, the total light production efficiency of a particle of atomic number Z and velocity v is determined solely by the energy deposit at distances from the track which exceed

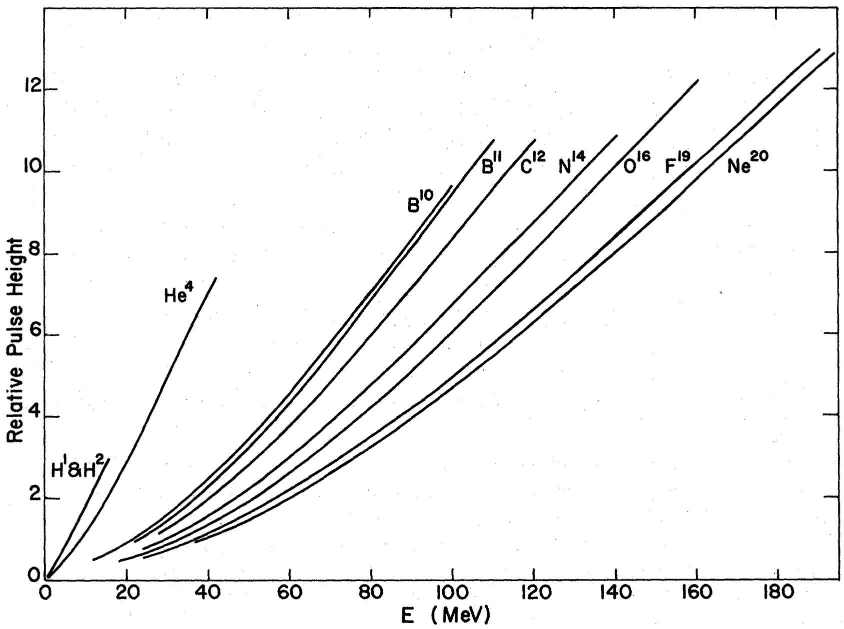

FIG. 1. Relative pulse height versus energy for heavy-ion bombardment of NaI(Tl) (Ref. 1).

the cylinder radius $R_c(Z, v)$. This is the energy available for luminescence, and is found to comprise a fraction of the total energy loss which decreases with decreasing particle velocity.

The present model involves two adjustable parameters, B and C. The former determines the magnitude of the cylinder radius for a given Z and v, while the latter functions as a normalization constant. Evaluation of B and C from a comparison of the model with the heavy-ion data of Newman and Steigert[1] is presented in the paper. Also presented is a calculation providing an approximate magnitude for ρ_c, the so-called "critical" density which separates the two regions of the crystal. In addition, a brief discussion is made regarding interpretation of the heavy-ion pulse-height characteristics of pure alkali halides at low temperature,[6] and those of anthracene and NE 102 plastic scintillators,[7] in terms of the present model.

II. PREVIOUS TREATMENTS

Theoretical analyses of the luminescent response of NaI(Tl) scintillation counters concern scintillation efficiency dL/dE. The most extensive treatment is the highly enlightening two-part analysis of Murray and Meyer.[2,3] These investigators demonstrate[2] that for electrons, protons, and He^4 ions, scintillation efficiency appears to depend only on the specific energy loss dE/dx. Further, they show that the Newman-Steigert data for heavy ions, plotted in the form dL/dE vs dE/dx as shown in Fig. 2, aggregate about the so-called "universal curve" (see Fig. 3). Indeed, Murray and Meyer make the explicit assumption that scintillation efficiency is a universal function of specific energy loss, and characterize the additional dependence on particle identity observed for heavy ions as a kind of "fine structure"—a detail to be considered separately.

In the first part of their analysis, only the universal curve is treated. Therefore, only that particular feature of the heavy-ion data is treated which is approximated by the universal curve, namely, the falling off of scintillation efficiency with increasing dE/dx. Murray and Meyer note that the characteristic room-temperature luminescence of NaI(Tl) is attributed to an electronic excitation of the Tl^+ ion. Also it is argued that the observed pulse-height magnitudes cannot be accounted for in terms of direct excitation by the penetrating ions, in view of the relatively small cross section for such events. Instead, one must assume a two-step process in which energy is transported from the particle track and subsequently absorbed at the activator centers. The authors regard exciton diffusion from the particle track as the dominant mode of energy transport ultimately leading to thallium luminescence. The excitons are assumed to result from the recombination of initially free electrons and holes. Further, they attribute the falling off of scintillation efficiency with increasing dE/dx to a saturation of available activator sites in regions of high exciton density. It should be noted that dE/dx is the key parameter in this formulation, uniquely determining probabilities for exciton formation, activator saturation, etc. This is a consequence of the assumption that all ionizations and subsequent recombinations occur directly

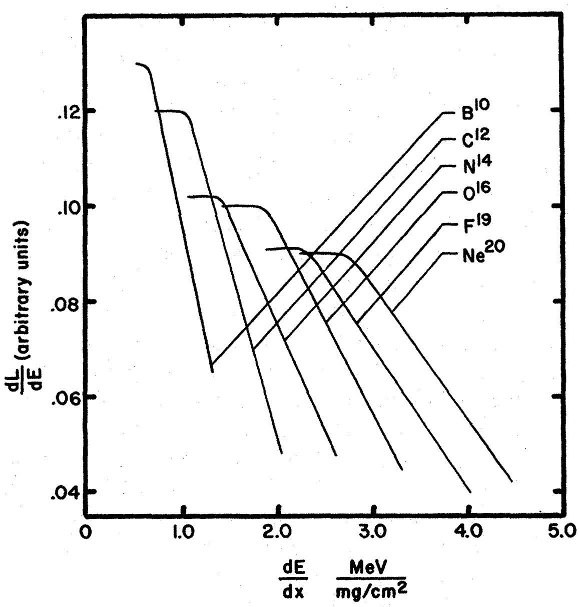

FIG. 2. Scintillation efficiency as a function of specific energy loss for NaI(Tl) (Ref. 1).

along the particle track, producing a line source of excitons with a density determined by the total specific energy loss. The thallium excitation probability is arrived at from simultaneous solution of equations representing exciton diffusion and activator saturation, subject to the initial exciton distribution and hence determined by dE/dx. The role played by dE/dx is diminished in importance when the so-called "track effect" is taken into account. As will be seen in the discussion of the present treatment, there is ample motivation to make such a refinement.

In the second part of their analysis,[3] Murray and Meyer treat the dependence on particle identity observed for heavy ions. This is accomplished by partitioning scintillation efficiency into two contributions: a part which is associated with light emission close to the particle track, and that attributed to δ rays—energetic secondary electrons which travel out beyond a fixed distance from the track, and into regions of the crystal where saturation effects are minimal. The pulse-height response to δ rays is assumed to be linear. Their effect on scintillation efficiency is determined by the fraction of the total dE/dx which they carry off. The latter quantity depends, at fixed dE/dx, on the charge of the penetrating ion. Thus, through δ rays, the dependence on particle identity is taken into account.

While the treatment of Murray and Meyer is quite successful in accounting for the pulse-height characteristics of NaI(Tl) and CsI(Tl) scintillation counters, it nevertheless follows from subsequent work that the validity of several of the basic assumptions made in the first part of their analysis is in doubt. (It is to be noted that, in contrast, the δ-ray model stands independent of prior assumptions. Further discussion of the δ-ray work, and comparison with the present formulation, is made in Sec. VII.) First, in a report by Gwin and Murray,[8] the activator-depletion hypothesis is examined in detail. It is shown that the shape of the dL/dE-vs-dE/dx curves for CsI(Tl) counters is independent of activator concentration, over several orders of magnitude variation of the latter. On the basis of this observation, the authors conclude that the behavior of scintillation efficiency at high $dE/$

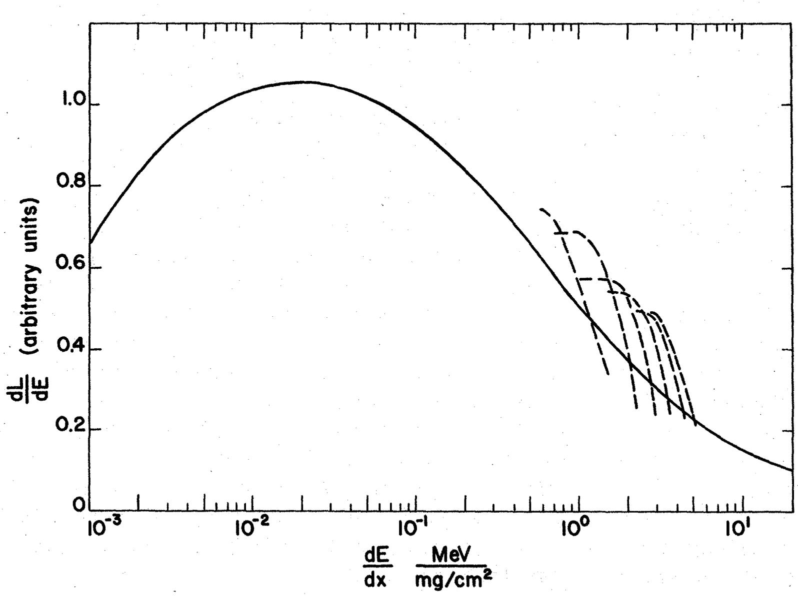

FIG. 3. Solid curve represents theoretical scintillation efficiency as a universal function of specific energy loss for NaI(Tl) (Ref. 2.) The dashed curves superimposed are the corresponding heavy-ion data of Ref. 1.

dx is not determined by a depletion of available activator sites. Rather, they suggest that the behavior is attributable to some intrinsic property of ionization density. This contention is supported by the observations of Blue and Liu[6] which indicate that dL/dE falls off with increasing dE/dx for pure alkali iodides in the same manner as in activated crystals.

Secondly, in view of much recent work reported on the nature of hole migration and electron-hole recombination in pure and impurity-activated alkali halides,[9–13] considerable doubt has also been shed regarding the assumption that exciton diffusion is the dominant mode of energy transport leading to activator luminescence. In particular, it has been amply demonstrated that ionizing radiation incident on pure alkali halides at low temperature creates initially free electrons, and holes which take on a self-trapped "V_k" configuration. The latter are generally mobile at temperatures well below room temperature, and migrate through the crystal by means of reorientation steps.[10] The recombination of an electron and hole results in a (V_k + electron) state, referred to as a "self-trapped exciton."[14] Polarized luminescence measurements at low temperatures[11] indicate that the self-trapped exciton does not reorient during its entire lifetime, and hence does not migrate through the crystal. At room temperature one might anticipate a somewhat greater mobility. However, it has been suggested[15] that this would be counteracted by the effect of a reduced lifetime, and thus a reduced diffusion length, which might be expected in view of the nonradiative decay mode of the self-trapped exciton at room temperature. It is therefore doubtful that the exciton serves as the major vehicle for energy transport.

It is important to note that while both the activator-depletion hypothesis and the assumption regarding the role played by excitons are of doubtful validity, in contrast, a third major feature of the Murray-Meyer formulation appears to stand on firm ground. In particular, the authors demonstrate that, at fixed dE/dx, the dependence of scintillation efficiency on particle identity is explained only when the track effect is taken into account. It is this insight which forms the basis of the present model.

A brief discussion is in order regarding a more recent attempt to explain the heavy-ion data. In this work, Katz and Kobetich[4] take detailed account of the track effect. They do so with the aid of energy-deposition profiles, each of which represents the density of deposited energy versus distance from the track for particles traveling in NaI at a particular velocity v. Curves are generated, on the basis of a semiempirical formulation developed by the authors,[16] for a set of velocities spanning the full range of the data. The energy-deposition profiles are used to establish the total dose of energy deposited in a single, fixed volume element associated with each of the Tl^+ ions. The authors adopt the previously held view of Murray and Meyer[2] that the nonlinearity of the response curves is attributed to depletion of available activator sites in regions of high ionization density. Thus, they assume that each thallium site can be excited only once subsequent to the passage of an incident particle. The excitation probability for a given thallium site is determined, through Poisson statistics for a one-hit process, by the dose of energy deposit inside the sensitive region surrounding the site, and an unknown critical dose E_0 corresponding to 63% probability for excitation. Reasonable agreement with the heavy-ion data of Newman and Steigert[1] is obtained for a value of E_0 equal to 4×10^7 erg/cm^3.

This author is in complete agreement with Katz and Kobetich regarding the need to take into account details of the spatial distribution of energy deposit when treating scintillation efficiency. However, it is believed, in view of the work of Gwin and Murray[8] and of Blue and Liu,[6] that thallium depletion is not an important factor in determining luminescent response, and should not be invoked as the dominant mechanism responsible for the falling off of scintillation efficiency.

III. ADDITIONAL INSIGHTS

Delbecq *et al.*[17] studied low-temperature recombination luminescence in KCl(Tl), focusing attention on the role played by the Tl^+ centers. They observed that subsequent to irradiation, stable Tl^0 and Tl^{++} centers are formed. Further, from their measurements of stimulated emission, they conclude that each of the following processes results in the characteristic thallium luminescence of the crystal:

(a) (Tl^+ + electron) $\to Tl^0$ followed by
(Tl^0 + hole) $\to (Tl^+)^* \to$ (Tl^+ + photon);

(b) (Tl^+ + hole) $\to Tl^{++}$ followed by
(Tl^{++} + electron) $\to (Tl^+)^* \to$ (Tl^+ + photon).

Of course, the fact that such sequential-capture processes can occur does not, in itself, imply that they constitute important luminescence mechanisms. In this regard, a key consideration is V_k mobility, which must be great enough to allow the V_k centers to move out from their point of creation and reach thallium sites in a time less than the lifetime of the luminescence. This question was explored by Dietrich and Murray in a recent diffusion calculation,[18] the results of which indicate that V_k diffusion lengths at room temperature are substantially greater than was previously anticipated[2] and, indeed, that separate V_k migration might be expected to play an important role in thallium luminescence.

IV. SUMMARY OF PAST FINDINGS

In summary, the following have been established, with varying degrees of certainty, regarding the heavy-ion response of thallium-activated alkali iodides:

(a) Nonlinearity of the response is not a consequence of activator depletion, but is attributed instead to some intrinsic property of ionization density

(b) Dependence of dL/dE at fixed dE/dx on the identity of the incident particle is related to the spatial distribution of energy deposit about the particle track.

(c) Sequential captures of electrons and holes at Tl^+ sites produce the characteristic thallium luminescence, and probably constitute the dominant luminescence mechanisms.

V. PRESENT FORMULATION

A. Parametrization

The heavy-ion data exhibited in Fig. 2 indicate clearly that dL/dE is not uniquely determined by dE/dx. Quantitatively, a comparison of the C^{12} curve with that for O^{16} shows that at fixed dE/dx scintillation efficiency varies by as much as a factor of 2. In contrast, consider a plot of these same data in the form dL/dE vs v (particle velocity) as shown in Fig. 4. These curves are derived by tracing the data reduction back one step. From Fig. 4 it is seen that at fixed velocity dL/dE varies across the data by the relatively small factor of $\frac{13}{9}$. It is interesting to note the corresponding variation in dE/dx. The latter can be obtained from the familiar stopping power relation

$$\frac{dE}{dx} \propto \frac{Z^2}{v^2} , \tag{1}$$

displaying the leading terms which characterize the slowing down of high-velocity ions of atomic number Z.[19] From Eq. (1) it is seen that the corresponding variation in dE/dx at fixed velocity is a factor of 4. Thus, at fixed velocity, scintillation efficiency is fairly insensitive to changes in stopping power. Combining this with the results of a similar analysis of Fig. 2, it is concluded that scintillation efficiency is far more sensitive to changes in velocity at fixed dE/dx than to changes in dE/dx at fixed velocity; i. e., dL/dE follows the variations in velocity more closely than those in dE/dx. Thus, velocity would appear the better of the two parameters in terms of which to describe scintillation efficiency.

The above conclusion has a rather interesting implication . First, let us note that the maximum energy which can be transferred by an incident particle to an electron in a classical head-on collision is given by

$$T_{\max} = 2mv^2 , \tag{2}$$

where m is the mass of the electron and v is the particle velocity. Further, from classical dynamics it can be shown that the dependence of energy transfer on the angle θ at which an electron is ejected from the particle track is given by

$$T = T_{\max} \cos^2 \theta . \tag{3}$$

This relation, together with range-energy data for electrons in the particular medium, determines the

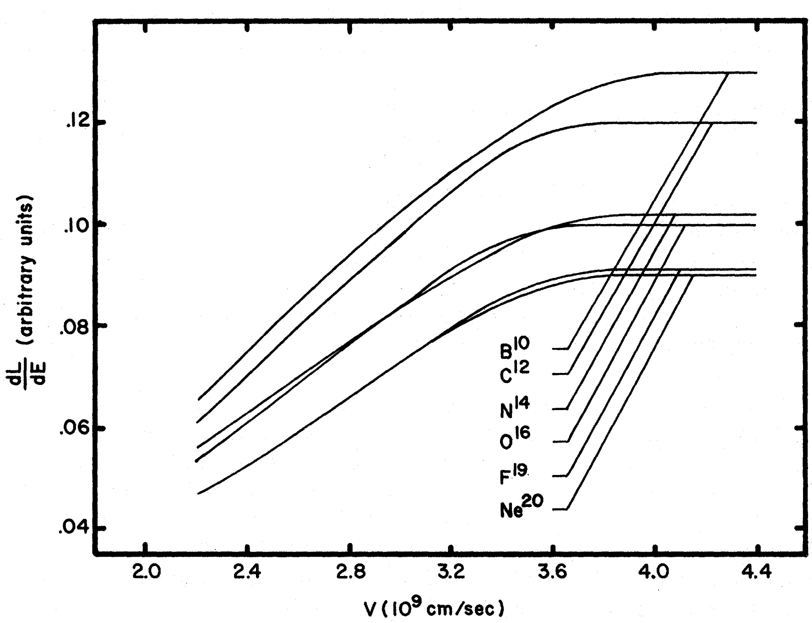

FIG. 4. Heavy-ion pulse-height data of Ref. 1 plotted as scintillation efficiency versus particle velocity.

spatial extent of energy deposit about the particle track. The latter is dependent, therefore, only on one parameter of the incident particle, namely, the velocity v. Thus, with regard to the relative sensitivity of dL/dE, one might state the following: Scintillation efficiency is less sensitive to changes in the total energy deposit (per unit path length) than to changes in its spatial distribution. It is therefore apparent that the track effect ought to play a central role in any theoretical account of the behavior of scintillation efficiency.

B. Model

Consider the penetration of an ionizing particle through an activated alkali halide crystal at room temperature. During the passage, free electrons and holes emanate from the particle track. Ionization cascades follow promptly until the region about the track, extending out perhaps several thousand angstroms, is a virtual sea of free, thermalized electrons, holes of the V_k configuration, and various other forms of lattice excitation. Subsequent events might be characterized as follows:

(i) *Luminescence processes*: processes which result in the emission of light from the crystal characteristic of the activator centers.

(ii) *Competitive processes*: processes which consume lattice excitation energy but do not result in the characteristic luminescence emission from the crystal.

Let us consider the so-called "competitive processes." Indeed, these are many in number and varied in character. Consequently, the problem of accounting for details of the heavy-ion pulse-height characteristics in terms of the deexcitation mechanisms operative in the crystal is highly complex and perhaps intractable. However, several of the more important competitive processes have one unifying feature: They are favored by high energy-deposit density and, consequently, are most important close to the particle track. The following is intended to illustrate this point.

First, competitive with sequential capture of electrons and holes at activator centers is the process of electron-hole recombination at normal lattice sites. The recombination rate goes approximately according to second-order kinetics in the energy-deposit density, and is therefore most prevalent in the high-density region close to the particle track. Secondly, to the extent that exciton migration and subsequent capture at activator sites is operative as a luminescence mechanism, one could classify exciton-exciton annihilation as a competitive process, also occurring with a rate proportional to the square of the energy-deposit density. (These nonlinear processes surely must relate to the observed nonlinearity of the pulse-height response.) Thirdly, one ought to consider the highly nonlinear lattice interaction processes. In particular, coloration of alkali halide crystals subsequent to ion bombardment has been observed to occur quite readily in the course of channeling experiments conducted at Chalk River,[20] and is accompanied by a rapid and drastic reduction in the channeling yield. This implies the creation of radiation-induced defect centers. Further, defect formation in alkali halides derives, in large part, from purely ionizing events as is demonstrated, e.g., by the recent study of Butterworth *et al.*[21] concerning the formation of color centers in KCl(Tl) by γ irradiation at room temperature. Thus, the production of defects is favored by high ionization density. The defect centers serve as electron and hole traps and therefore render some of the lattice excitation energy unavailable for luminescence. In addition, it has been suggested[15] that transient color center production might be expected to result in internal absorption of photons emitted by the activator ions, thus rendering the crystal partially opaque to the activator emission. In short, radiation damage effects are competitive with luminescence processes, and are favored by high energy-deposit density. Finally, and perhaps of greatest importance, are effects associated with lattice heating in the vicinity of the track of a highly ionizing particle. In particular, it is well known that the radiative decay probability of the activator centers, in, e.g., NaI(Tl),[22] falls off rapidly with increasing temperature. One might therefore expect a similar effect close to the track of a penetrating ion. Reduction of the radiative decay probability is competitive with the luminescence processes, and is favored by high energy-deposit density, as are the various other competitive processes mentioned above.

In view of this unifying feature, a gross simplification of the problem can be effected. In particular, the present formulation employs the concept of a cylinder surrounding the particle track, inside of which the density of deposited energy exceeds some critical value ρ_c. The crystal is thus partitioned into regions of high and low energy-deposit density. From each region there is a corresponding contribution to the total scintillation efficiency. The latter is expressed as

$$\frac{dL}{dE} = \left(\frac{dL}{dE}\right)_i F_i + \left(\frac{dL}{dE}\right)_o F_o \,, \tag{4}$$

where i and o refer, respectively, to regions inside and outside the high-density cylinder, and where F_i and F_o are the corresponding fractions of the total energy deposit per unit path length. The assumption is made that competitive processes dominate close to the particle track, and render the contribution to dL/dE from inside the cylinder a negligible fraction of the total, i.e.,

$$\left(\frac{dL}{dE}\right)_i F_i \ll \left(\frac{dL}{dE}\right)_o F_o \, . \qquad (5)$$

Further, it is assumed that the response to energy deposited outside the high-density cylinder is linear, i.e.,

$$\left(\frac{dL}{dE}\right)_o = \text{const.} \qquad (6)$$

The latter assumption is motivated by the observation that the response to more weakly ionizing particles, e.g., energetic protons, is indeed linear. The total scintillation efficiency is then given by

$$\left(\frac{dL}{dE}\right) = C_0 F_o(Z, v) \, , \qquad (7)$$

where C_0 is a constant and F_o is a function of the atomic number and velocity of the incident particle. F_o is interpreted as the fraction of the total energy deposit available for luminescence, and will be seen to exhibit the same general dependence on charge and velocity as do the heavy-ion pulse-height data.[1]

Thus, in the present formulation, the problem reduces to one involving only energy-deposit considerations. Explicit assumptions regarding luminescence mechanisms and competitive processes are not made. Rather, only the one unifying feature, namely, the enhancement of competitive processes by high ionization density, is used in order to account for the general character of the heavy-ion data.

C. Details

By definition,

$$F_o(Z, v) = \left(\frac{dE}{dx}\right)_0 \Big/ \left(\frac{dE}{dx}\right)_{\text{tot}} \, , \qquad (8)$$

where $(dE/dx)_0$ is expressed in terms of the density of energy deposit as follows:

$$\left(\frac{dE}{dx}\right)_0 = \int_{R_c(Z,v)}^{R_{\max}(v)} \rho(Z, v, R) \, 2\pi \, R dR \, . \qquad (9)$$

In Eq. (9), $\rho(Z, v, R)$ is the density of deposited energy at a distance R from the track of a particle of atomic number Z traveling with velocity v. $R_{\max}(v)$ is the maximum distance from the track at which energy is deposited. $R_c(Z, v)$ is the charge- and velocity-dependent cylinder radius.

1. $\rho(Z, v, R)$

The calculations of Katz and Kobetich[4,16] provide numerical estimates of the energy-deposition function. However, simplifying assumptions are incorporated which render their precise numerical values somewhat uncertain. For example, it is assumed that all electrons ejected by an ionizing particle travel out at right angles from the track. This assumption surely results in an overestimate of the spatial extent of the energy deposit, i.e., an overestimate of $R_{\max}(v)$, since the more energetic electrons tend to move, at least initially, in the forward direction. Qualitatively, their calculations indicate that for distances R not too close to the particle track ($R \gtrsim 100$ Å),

$$\rho(Z, v, R) \propto Z^{*2}/R^2 , \qquad (10)$$

where Z^* is the effective charge of the incident particle, defined as the ratio of the stopping power of the particle to the stopping power of a proton traveling at the same velocity and given by

$$Z^* = Z\,(1 - e^{-125\beta Z^{-2/3}}) , \qquad (11)$$

in which β is the ratio v/c c being the velocity of light in vacuum. Further, the energy-deposition profiles for NaI indicate an additional velocity dependence differing little from a multiplicative factor of $1/v^2$. Thus, for distances not too close to the particle track, the numerical results of the calculations of Katz and Kobetich can be approximated by the relation

$$\rho(Z,v,R) = kZ^{*2}/R^2 v^2 , \qquad (12)$$

where k is a constant. The above expression is regarded as adequate for present purposes, and is employed in the integral of Eq. (9) to calculate $F_o(Z,v)$.

2. $R_c(Z, v)$

The high-density cylinder is defined so that over the surface (i.e., at $R = R_c$) ρ equals ρ_c, an undetermined, critical value for the density of deposited energy. Thus, from Eq. (12) one obtains

$$R_c(Z,v) = B\ Z^*/v , \qquad (13)$$

where B is a constant equal to $(k/\rho_c)^{1/2}$. Since the dependence of Z^* on v for high-velocity ions is fairly weak, it follows from Eq. (13) that the cylinder radius increases with decreasing velocity. This is a major contributing factor to the subsequent decrease of F_o and the corresponding falloff of scintillation efficiency as the particle slows down. Further, note that it is the value of B, and therefore the magnitude of the cylinder radius, which results from a fit of the model to data, and not the value of the critical density ρ_c. An estimate of the latter may be obtained by normalizing the energy deposit to $(dE/dx)_{\text{tot}}$ and thus solving for the constant k in the energy-deposition function. However, this can be accomplished only upon making some assumption regarding the form of ρ close to the particle track. This is done in Sec. VI.

3. $R_{max}(v)$

The maximum distance from the particle track at which energy is deposited is taken as the maximum transverse component of the practical range of secondary electrons. In particular, from practical-range-vs-energy data for electrons in various media (Al, Ag, Au) Meyer and Murray[3] perform an extrapolation to obtain the practical range R_p in NaI as $0.012T^{1.35}$ mg/cm^2, where T is the energy of the secondary electron in keV. Combining this with Eqs. (2) and (3), the practical range as a function of the angle θ at which an electron is ejected from the particle track is given by

$$R_p(\theta) = 0.012T_{max}^{1.35}\cos^{2.70}\theta\,. \tag{14}$$

The transverse component, $R_p(\theta)\sin\theta$ attains a maximum value at $\theta \approx 31°$. Thus $R_{max}(v)$, expressed in terms of velocity, is given by

$$R_{max}(v) = 1.32\times 10^{-6}(v/v_r)^{2.70}\text{ cm}, \tag{15}$$

where v_r is a reference velocity equal to 10^9 cm/sec. Typical values for $R_{max}(v)$ corresponding to the heavy ions of the Newman-Steigert data[1] are less than those obtained by Katz and Kobetich[4] by a factor of 1/3, and range from 1000 to 7000 Å, decreasing fairly rapidly with decreasing velocity. Indeed, with decreasing particle velocity, R_{max} and R_c approach each other and, consequently, dL/dE approaches zero. This places a lower bound on velocity for applicability of the model.

4. $(dE/dx)_{tot}$

Total stopping power is represented by the expression[19]

$$\left(\frac{dE}{dx}\right)_{tot} = \frac{4\pi Z^{*2}e^4}{mv^2}\, n\ln\frac{2mv^2}{I}\,, \tag{16}$$

which is equivalent to Bethe's formulation[23] for the stopping of high-velocity protons, modified by an effective charge Z^* so as to be made applicable to heavy ions. The parameter n is the average electron density of the medium, and I is the mean excitation energy, evaluated by Sternheimer for NaI to be 0.427 keV.[24] Heavy-ion stopping-power curves generated from Eq. (16) are displayed in Fig. 5. These agree with the corresponding semiempirical curves generated by Newman and Steigert[1] to within several percent for velocities $v \gtrsim 2.2\times 10^9$ cm/sec, but rise more rapidly than do the latter at lower velocities. The present calculation is confined to the high-velocity region in which the analytic expression of Eq. (16) is regarded as adequate and, indeed, preferable to the empirical curves for calculation purposes.

Employing the relations of this section in Eq. (7), scintillation efficiency becomes

$$\frac{dL}{dE} = C\,\frac{\ln[1320(v/v_r)^{3.70}(BZ^*)^{-1}]}{\ln[2.67(v/v_r)^2]}\,, \tag{17}$$

which constitutes the result of the model in the present approximation.

VI. COMPARISON WITH EXPERIMENT

In order to evaluate the undetermined parameter B and the appropriate normalization constant for

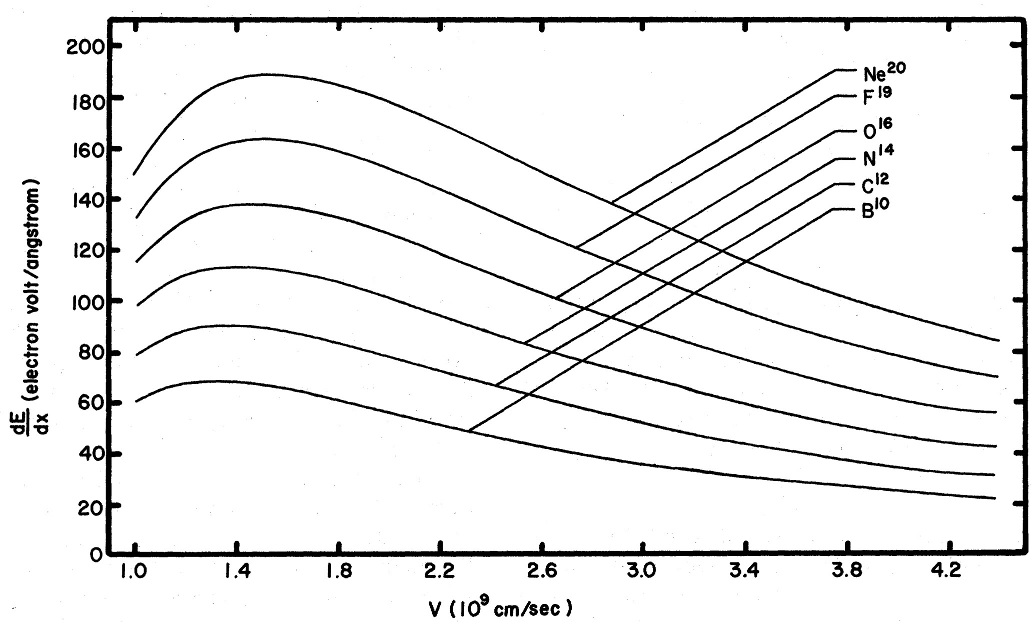

FIG. 5. Calculated values of specific energy loss as a function of particle velocity for heavy-ion penetration of NaI.

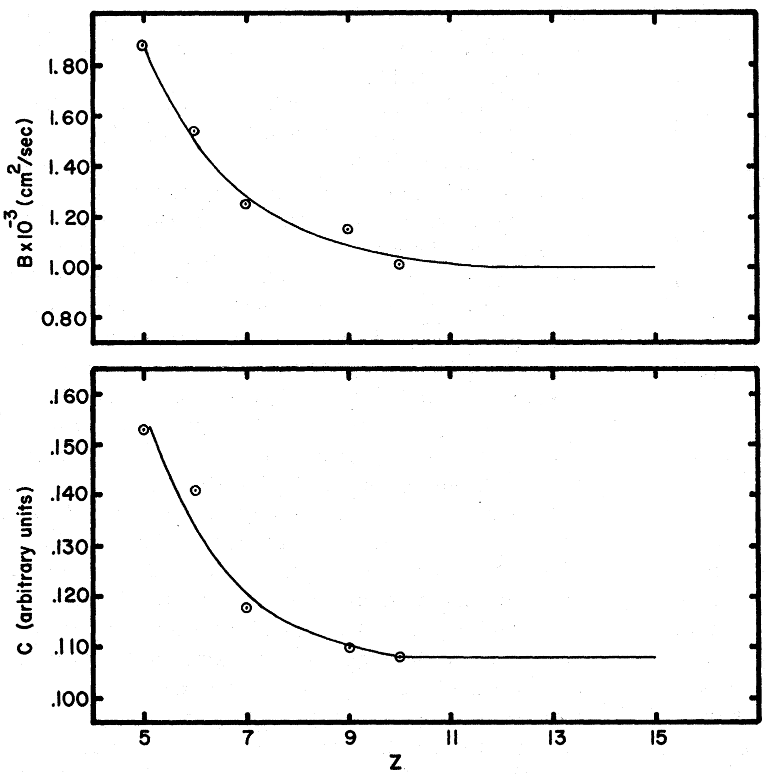

FIG. 6. Values of the parameter B and normalization constant C generated from a fit of each of the experimental curves in Fig. 4 at two points ($v = 2.5 \times 10^9$ cm/sec and $v = 4.0 \times 10^9$ cm/sec).

the Newman-Steigert data, each curve, in turn, is fitted at two points ($v = 2.5 \times 10^9$ cm/sec and $v = 4.0 \times 10^9$ cm/sec). The resulting values for B and C are displayed in Fig. 6. Values for O^{16} are anomalously low in consequence of experimental uncertainty in the response curve (note the exaggerated curvature of the O^{16} data in Fig. 4) and are omitted. Clearly, a unique value for B which would provide a fit across the entire set of data does not result. However, it is indicated that B and C each approach asymptotic values with increasing Z. This implies that a unique parametrization does indeed exist for high-Z data, i.e., for response curves corresponding to $Z \gtrsim 10$. Further, it is noted that the asympotic value of B is fairly insensitive to the particular choice of velocities in the data fit, provided that at least one of the values is taken from the nonlinear region.

Evaluation of dL/dE in Eq. (17) by inserting B (asymptotic) equal to $1.00 \times 10^{+3}$ cm^2/sec and normalizing to the Ne^{20} curve of the heavy-ion data at v equal to 2.5×10^9 cm/sec results in the set of theoretical curves displayed in Fig. 7. Clearly, the two main features of the data are qualitatively accounted for, namely, the falling off of scintillation efficiency with decreasing velocity and the charge dependence at fixed velocity. For purposes of closer comparison, the theoretical curves for B^{10}, N^{14}, and Ne^{20} are superimposed on the corresponding data in Fig. 8. There is reasonable agreement between theory and experiment for the two heavier ions over the entire range of velocities, but agreement with the B^{10} data only at the low-velocity end. The regions of best agreement correspond to portions of the particle trajectories over which lattice excitation is greatest.

Several additional points ought to be noted. First, as seen in Fig. 4, the data become linear at a

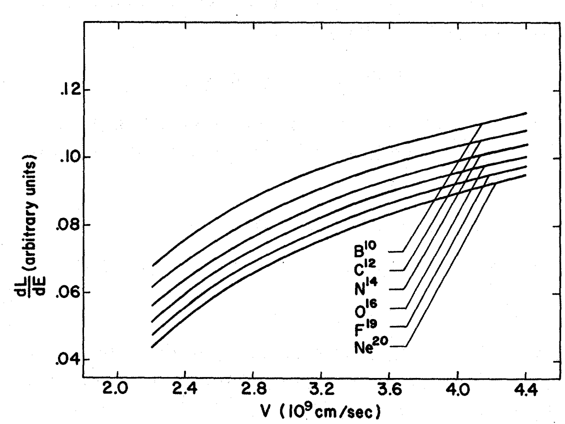

FIG. 7. Calculated values of scintillation efficiency versus particle velocity for heavy-ion bombardment of NaI(Tl).

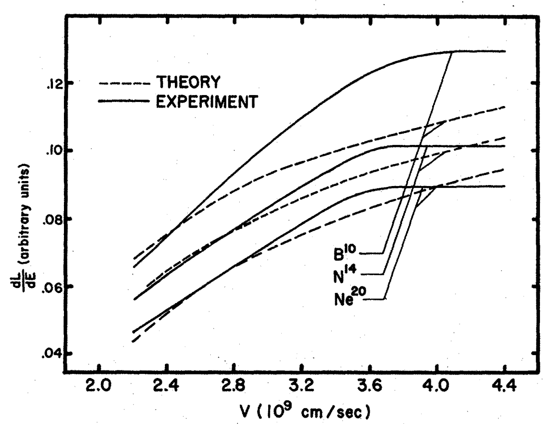

FIG. 8. Comparison of theory with experiment for three heavy ions in NaI(Tl).

velocity of approximately 3.8×10^9 cm/sec for most cases (slightly higher for B^{10}). However, in view of errors resulting from reduction of the data, and since no correlation between that particular velocity and electronic processes in the crystal is apparent, one might doubt whether scintillation efficiency does indeed become linear. This question was raised previously,[8] but not examined for heavy ions. Thus, no attempt is made in the present formulation to account for the linearity of the data. Similarly, the apparent pairing of the curves in Fig. 4 is not treated; nor is it believed to be a real effect. In particular, it is difficult to see why, e.g., bombardment with high-velocity N^{14} and O^{16} ions, each essentially stripped of all electronic charge, should produce almost identical results, while the response to C^{12} is so very different. Finally, the precise shape of the theoretical curves is somewhat sensitive to the mathematical form of the energy-density function assumed in Eq. (12). However, as mentioned previously, the latter is regarded as being adequate for present purposes.

Estimation of ρ_c and F_o

The asymptotic value of B yields a range of values for the radius of the high-density cylinder given by

$$110 \text{ Å} \lesssim R_c\,(Z, v) \lesssim 390 \text{ Å}, \tag{18}$$

while the corresponding range of $R_{max}\,(v)$ is

$$1100 \text{ Å} \lesssim R_{max}\,(v) \lesssim 7200 \text{ Å}. \tag{19}$$

Comparing, it is apparent that the dominance of high-density effects is restricted to regions relatively close to the particle track, i.e., close as compared to R_{max}. Further, in view of the sharp cutoff nature of the model, the values for R_c might be regarded as providing upper bounds on the region over which competitive effects truly dominate. In this sense, the range of values shown in Eq. (18) appears reasonable.

Additional insight regarding the validity of the results of the present formulation might be provided by an estimate of the critical energy density ρ_c and the range of values for F_o. However, as mentioned previously, this requires knowledge of the density of deposited energy all the way down to the particle track. Such information is not available; nor is it easily acquired. For purposes of obtaining a rough idea of the magnitudes of ρ_c and F_o, the following approximation is made: ρ is assumed to fall off as $1/R^2$ from some distance $a(v)$ out to R_{max}, while over the region $R \lesssim a(v)$ the density is assumed to be constant. This distribution is shown schematically in Fig. 9.

Indeed, a leveling off of ρ at small R is indicated by the results of the calculations of Katz and Kobetich,[4] although those calculations do not extend down to zero distance from the track. Further, one might anticipate a leveling off in view of electron crossover of the track of a positive ion. The crossover region is limited to the maximum distance for direct influence of the incident particle. Consequently, the cutoff distance $a(v)$ corresponding to the distribution in Fig. 9 is taken as the distance at which the field of the incident particle is just canceled by the induced polarization field of the electrons in the medium. This value is readily obtained from classical theory of an electron gas, and results in the following:

$$a(b) = v/\omega_0 , \tag{20}$$

where ω_0 is the natural plasma frequency $(4\pi ne^2/m)^{1/2}$ of a gas of density n. For NaI, $a(v)$ varies from 4 to 8 Å over the velocity range of the heavy-ion data.

Employing the distribution of Fig. 9 in an integration to obtain the total energy deposit per unit path length, and equating to $(dE/dx)_{tot}$ in Eq. (16), an expression for the parameter k in the energy-deposition function is obtained. The latter exhibits

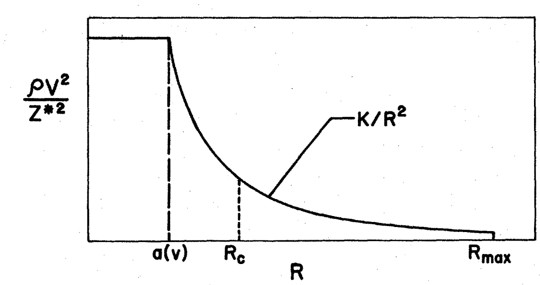

FIG. 9. Simplified model for the dependence of energy-deposit density on atomic number, velocity, and distance from the track of a penetrating ion.

a weak velocity dependence which, when averaged over the range of the data, yields k equal to 5.32×10^{13} erg cm/sec^2. Thus, k can now be separated from ρ_c and the latter takes on the value

$$\rho_c = 5.32\times 10^7 \text{ erg/cm}^3, \tag{21}$$

which corresponds, roughly, to one ionization per luminescence center. Such a density is required for the onset of competitive processes. Thus, with regard to ρ_c, the model appears to be self-consistent.

The range of values for F_o corresponding to the heavy-ion data is given by

$$0.20 \lesssim F_o \lesssim 0.50. \tag{22}$$

While these values are only approximate, they do indeed indicate that enough energy is deposited outside the high-density cylinder to account for the total light production.

VII. COMPARISON WITH δ-RAY MODEL

The present model bears certain distinct similarities in underlying concept and mathematical form to the δ-ray work of Meyer and Murray.[3] In view of this, a detailed comparison is in order.

Each formulation is based on the premise that light production efficiency is a decreasing function of energy-deposit density. The problem is made tractable by assuming a partitioning of the crystal about the particle track into regions of high and low density associated, respectively, with regions inside and outside a critical cylinder. The essential differences between the two models lie in the manner in which this partitioning is effected, the assumptions regarding the contributions to the total light output from each region, the parametrization, and the method of calculation.

The δ-ray formulation assumes a cylinder of fixed radius. It might be argued that this manner of partitioning is somewhat unrealistic, in view of the rather large variations in details of the energy-deposit density along the trajectories of the particles treated. In particular, corresponding to the velocity range of the heavy-ion data,[1] the spatial extent of energy deposit (as given by R_{max}) varies approximately by a factor of 7, while the total rate of energy loss varies by more than a factor of 2. Further, the assumption of a fixed cylinder radius results in values for F_o which, at fixed velocity, vary by as much as 20–25% across the heavy-ion data. This appears to be incompatible with what one might anticipate on the basis of energy-deposit theory. In particular, from Eqs. (8), (9), (12), and (16) it is seen that at fixed velocity the fraction of the total energy deposit outside a fixed R_c is independent of the identity of the particle. In contrast, the present formulation assumes a partitioning on the basis of a critical value of energy-deposit density, which this author regards as a relatively natural and direct way to distinguish between the two regions, and results in a charge- and velocity-dependent cylinder radius, and corresponding function F_o.

The δ-ray model treats $(dL/dE)_i$ as a continuous function of $(dE/dx)_i$. The present formulation circumvents the problem of treating $(dL/dE)_i$ by assuming that competitive processes render $(dL/dE)_i F_i$ insignificant when the cylinder density is high enough. The critical density ρ_c is contained in the undetermined parameter of the model.

The δ-ray model employs dE/dx as the independent variable, in contrast to the use of particle velocity in the present work. The latter provides a better parametrization of the heavy-ion data and greatly simplifies the calculations, although no apparent change in the physics of the problem is so introduced.

The resulting mathematical expression for dL/dE in the present formulation is a simple analytic form which more readily lends itself to application to other systems, and generalization to other domains of charge and velocity, than does the numerical, iterative procedure employed in the δ-ray model.

Finally, it should be emphasized that neither treatment is dependent on explicit assumptions regarding luminescence mechanisms or competitive processes.

VIII. DISCUSSION OF OTHER SYSTEMS

As indicated by the work of Blue and Liu,[6] scintillation efficiency for the entire family of pure alkali iodides subject to energetic-α-particle bombardment at low temperature falls off with decreasing particle velocity, while the response to protons is linear. This is precisely the same behavior as is exhibited by the Newman-Steigert data for NaI (Tl). However, the mechanisms responsible for the characteristic luminescence are undoubtedly different. Indeed, that which is the dominant mechanism in the pure-crystal case (electron-hole recombination) is most probably a major competitive mechanism in the latter. Similarly, the heavy-ion response of organic phosphors such as anthracene and NE 102 plastic scintillators[7] exhibits the same falling off of dL/dE with decreasing energy, and the same systematic increase in light output with decreasing atomic number at fixed energy, as is observed in NaI(Tl). Here again the luminescence mechanisms are quite different. It is tempting to attribute the behavior of heavy-ion scintillation efficiency to a single process common to all such scintillators. The present model suggests a possibility. In particular, it is suggested that the region close to the track of a highly ionizing particle penetrating through a phosphor is rendered relatively inefficient as a scintillating medium, in view of the dominance of competitive effects favored by high ionization density. The charge and velocity dependence of the response data is determined, in large part, by the behavior of the function $F_o(Z, v)$ [Eq. (8)] giving the fraction of the total energy deposit available for efficient light production.

IX. SUMMARY

The aims of the present work were twofold, namely, to provide a theoretical account of the response of NaI(Tl) to room-temperature bombardment by energetic heavy ions and, in so doing, to present a theory applicable to a fairly wide range of scintillating crystals. The present formulation is of a general nature, independent of precise details regarding luminescence mechanisms. It provides a qualitative account of the general character of the NaI(Tl) data,[1] and is in reasonable quantitative agreement with experiment, particularly for high-Z particles. The model appears to be self-consistent, as is indicated by the approximate values obtained for several of its parameters. Further refinement of the model at this time would be unwarranted, in view of relatively large uncertainties in existing data.

In addition to the account of the NaI(Tl) data, the present formulation provides a qualitative understanding of the heavy-ion response characteristics of, e.g., pure alkali iodides, anthracene, and plastic scintillators such as NE 102. In so doing, a common feature affecting the response of the various scintillators is uncovered, namely, the importance of high-ionization-density effects.

In order to test, further, the validity of the track-effect theory, it would be highly desirable to have available a detailed set of data corresponding to high-energy ($E/A \gtrsim 1$ MeV/nucleon) bombardment of, e.g., NaI(Tl) by ions of atomic number $Z \gtrsim 10$. Refinement of the model to provide a precise quantitative account of the data would entail, primarily, a corresponding refinement of the energy-deposition calculations of Katz and Kobetich.[4,16]

Note added in proof. Some of the main ideas of the present track-effect theory are contained in a previous paper by Myron Luntz and Ralph H. Bartram, Phys. Rev. 175, 468 (1968).

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