Stopping Power and Luminescent-Response Calculation for Channeling in NaI(Tl) and CsI(Tl)*

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Stopping powers of the most prominent axial channels in NaI(Tl) and CsI(Tl) are calculated for well-channeled ions (He^4–Ne^{20}) over the velocity range $E/A=1$–10 MeV/amu. An electron-gas model is applied to a treatment of the excitation of valence electrons, and an additional term is constructed which accounts for core-electron excitation. Nonparticipating electrons are excluded by application of an adiabaticity criterion. The channel-stopping powers that result are typically 20–25% of those characteristic of random incidence, indicating a relatively pronounced channeling effect for the alkali iodides. The calculated curves are combined with appropriate scintillation-efficiency data (dL/dE versus dE/dx) corresponding to bombardment of NaI(Tl) and CsI(Tl) scintillation counters, in order to obtain a prediction of the luminescent response (L versus E) to the channeled particles, and thus to account quantitatively for a reported anisotropy in the scintillation spectra. Higher than normal scintillation efficiency is predicted for the channeled particles, indicating that the crystal channels in NaI(Tl) and CsI(Tl) are axes preferential for luminescence. This effect may be explained qualitatively by reduced ionization density for channeled particles, which favors radiative recombination of electrons and holes at Tl^+ sites over competing nonradiative recombination.

I. INTRODUCTION

THE pulse-height characteristics of NaI(Tl)[1] and CsI(Tl)[2] scintillation counters, under energetic heavy-ion bombardment, have been studied in detail at Yale University. During the course of these and subsequent investigations,[3] observations were made that indicate a crystal-orientation dependence in the luminescent response of the counters. A precise experimental account of this anisotropy has not, as yet, been reported. A theoretical account is given in the present paper. The observed anisotropy can be explained qualitatively in terms of the *channeling* phenomenon; an effect that manifests itself in anomalously low stopping powers (dE/dx) of crystalline media for the slowing down of charged particles incident in major symmetry directions. Channeling is well established experimentally for both low[4,5] and high[6–12] velocities, and has been accounted for theoretically by Lindhard.[13,14] The relation between channeling and the observed luminescent-response anisotropy derives from the relatively low stopping power characteristic of the channel trajectory, and the manner in which stopping power determines the luminescence. The latter is exhibited by the Yale scintillation-efficiency curves of Fig. 1, which illustrate the dependence on stopping power of the scintillation efficiency, dL/dE, corresponding to bombardment of NaI(Tl)[1] and CsI(Tl)[2] with a given set of heavy ions. These curves were obtained from a combination of luminescent-response data, L versus E, and corresponding semiempirical stopping-power curves (dE/dx versus E) constructed to represent the specific energy loss of randomly incident heavy ions. The curves of Fig. 1 clearly indicate, for each case, a marked reduction in scintillation efficiency at relatively high stopping powers. In view of this behavior, and upon consideration of the anomalously low stopping power characteristic of the channel trajectory, it would appear to follow that a particle channeled in either of these phosphors produces light more efficiently than one randomly incident, and consequently induces a brighter response when the ion is completely stopped in the crystal. It is therefore postulated that the crystal channels in NaI(Tl) and CsI(Tl) are directions preferential for luminescence.[15]

The object of the present investigation is to treat theoretically the interactions between well-channeled particles and the constituents of thick NaI(Tl) and

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

[2] E. Newman, A. M. Smith, and F. E. Steigert, Phys. Rev. **122**, 1520 (1961).

[3] F. E. Steigert (private communication).

[4] G. R. Piercy, F. Brown, J. A. Davies, and M. McCargo, Phys. Rev. Letters **10**, 399 (1963).

[5] M. McCargo, J. A. Davies, and F. Brown, Can. J. Phys. **41**, 1231 (1963).

[6] G. Dearnaley, IEEE Trans. Nucl. Sci. **11**, 243 (1964).

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[8] W. M. Gibson, C. Erginsoy, H. E. Wegner, and B. R. Appleton Phys. Rev. Letters **15**, 357 (1965).

[9] B. R. Appleton, C. Erginsoy, H. E. Wegner, and W. M. Gibson Phys. Letters **19**, 185 (1965).

[10] B. R. Appleton, C. Erginsoy, and W. M. Gibson, Phys. Rev. **161**, 330 (1967).

[11] A. R. Sattler and G. Dearnaley, Phys. Rev. **161**, 244 (1967).

[12] A. R. Sattler and G. Dearnaley, Phys. Rev. Letters **15**, 59 (1965).

[13] J. Lindhard, Phys. Letters **12**, 126 (1964).

[14] J. Lindhard, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **34**, No. 14 (1965).

[15] This suggestion was first made by F. E. Steigert (private communication) subsequent to the Yale experiments.

CsI(Tl) single crystals, for the purpose of obtaining a prediction of the luminescent response to the channeled particles, and thus a quantitative account of its anisotropy. The particles treated are those corresponding to the Yale scintillation-efficiency data,[1,2] namely, He^4, B^{10}, B^{11}, C^{12}, N^{14}, O^{16}, F^{19}, and Ne^{20}, with energies $E/A = 1$–10 MeV/amu. The method used amounts to a reversal of the data-reduction procedure employed by the Yale group, and consequently involves the construction of channel stopping-power curves. The present treatment is limited to the case of axial channeling, and concerns only the *best* channeled particles, i.e., those particles that experience the smallest dE/dx. Trajectories of such particles are approximated to lie directly along the channel axes. The stopping-power calculation and the luminescent response predictions are treated in Secs. II and III, respectively. In Sec. IV a qualitative model for the reduction of dL/dE at high dE/dx and suggestions for further investigation are considered.

II. CHANNELING AND STOPPING-POWER CONSIDERATIONS

General Assumptions

All channeling and stopping-power considerations are made without regard to the thallium impurities. This is justifiable in terms of the low thallium concentrations of interest (typically 0.13 mole %),[1,2] and the substitutional nature of the doping.[16] Furthermore, all calculations are performed assuming a perfectly static lattice. This seems reasonable in light of recent indications that both the criteria for channeling[10] and the maximum range for *best* channeled particles[17] are highly insensitive to lattice temperature.

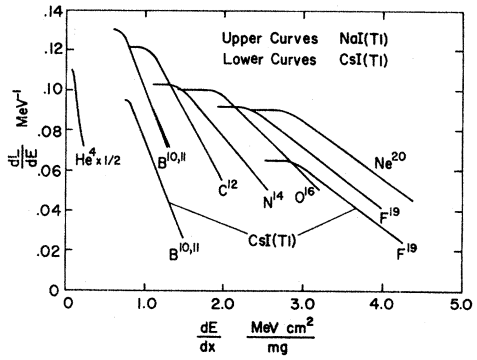

FIG. 1. Experimental scintillation efficiency as a function of specific energy loss for NaI(Tl) (Ref. 1) and for CsI(Tl) (Ref. 2).

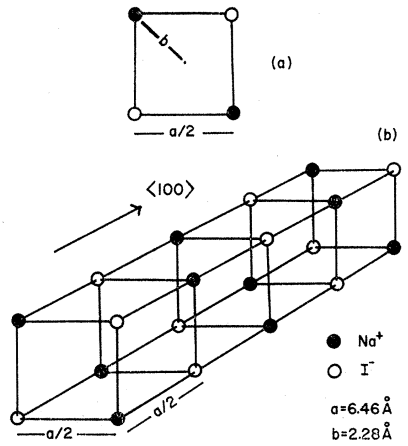

FIG. 2. Projection (a) and schematic representation (b) of the ⟨100⟩ axial channel in NaI.

Channel Identification

The most prominent axial channels lie between the most dense rows of atoms and, by inspection of the NaI and CsI crystal structures, are taken to be defined by the NaI⟨100⟩, NaI⟨110⟩, CsI⟨111⟩, and CsI⟨100⟩ axes. This identification is made without regard to the distinction between anions and cations. Such a distinction would be expected to affect the finer details of a channel trajectory, but not the identity of the channels themselves. A view of the NaI⟨100⟩ axial channel is shown in Fig. 2.

Acceptance Angles

Insight regarding the stopping powers to be expected of the various channels can be gained from a comparison of the respective angles of acceptance, or maximum allowed incidence angles for channeling, with measured acceptance angles for other cases, namely, for proton channeling in silicon and germanium.[10] The NaI and CsI acceptance angles were calculated in accordance with existing channeling theory,[13,14,18] generalized sufficiently to take into account the diatomic nature of the crystals. In particular, two channel types are encountered in the present application. One type is bounded by monatomic rows of both ionic species present in the crystal. For such channels, the acceptable angle is calculated from the lower of the two barrier potentials, where the latter are evaluated according to the methods of Ref. 18. The other type is bounded by identical rows with alternating ions. In these cases, the barrier potential used to calculate the acceptance angle is the string potential evaluated at the larger Thomas-Fermi radius, and is a sum of contributions from the two species

[16] F. Seitz, J. Chem. Phys. **6**, 150 (1938).
[17] L. Eriksson, Phys. Rev. **161**, 235 (1967).
[18] C. Erginsoy, Phys. Rev. Letters **15**, 360 (1965).

TABLE I. Acceptance angles for the most prominent axial channels in NaI and CsI.

Crystal	Channel	Acceptance angle[a] (deg)
NaI	⟨100⟩	$0.633(Z_i/E)^{1/2}$
	⟨110⟩	$0.387(Z_i/E)^{1/2}$
CsI	⟨111⟩	$0.922(Z_i/E)^{1/2}$
	⟨100⟩	$0.849(Z_i/E)^{1/2}$

[a] Energy E is in units of MeV.

of ions. Details of these calculations are presented elsewhere.[19] The results are displayed in Table I, in terms of the atomic number Z_i and energy E of the incident particle. Upon comparison one finds that the alkali-iodide channels have considerably larger acceptance angles than do comparable channels in silicon and germanium: for example, the acceptance angles of the alkali iodides for channeling of a 5-MeV proton range up to 0.413 deg, in comparison with a value of 0.2 deg for the silicon ⟨110⟩ axis, the latter having been measured by Appleton *et al.*[10] and calculated from the same relation as was employed in the present work. The comparative openness of the alkali-iodide channels leads one to anticipate relatively pronounced channeling effects, in particular, a relatively large reduction in the stopping power from that characteristic of random incidence. In silicon, the major axial channels correspond to a stopping power reduction by a factor of approximately $\frac{1}{2}$,[10] while in germanium the reduction factor has been observed to exceed $\frac{1}{3}$.[10,12] Larger reductions are anticipated for NaI and CsI.

Stopping Power Model

The alkali-iodide stopping powers for the various particle-channel combinations are obtained by appeal to the treatment of Appleton, Erginsoy, and Gibson,[10] in which an evaluation is made of the stopping power of thin, silicon single crystals for *best*-channeled, MeV-energy protons. With particle trajectories in the silicon crystals approximated to lie along the various channel axes (or mid-planes in cases of planar channeling), and upon application of an adiabaticity criterion, it was established that for the most open channels a proton of energy less than 3 MeV is capable of exciting only the valence electrons. It was then assumed that the *excitable* valence electrons respond to the incident particles in the manner of a free-electron gas. The stopping power of an electron gas was partitioned, according to the theory of Bohm and Pines,[20] into contributions due to individual-electron excitations and collective (plasma) excitations. The resulting stopping power for incident particles of charge Z_ie and velocity v is given by

$$-\frac{dE}{dx}=\frac{4\pi Z_i^2e^4}{mv^2}N\left(Z_{\text{loc}}\ln\frac{2mv}{\hbar k_c}+Z_{\text{val}}\ln\frac{k_c}{\omega_0/v}\right), \quad (1)$$

where N is the number of atoms per unit volume, and where NZ_{loc} and NZ_{val} are effective densities for one-electron excitations and plasma excitations, respectively. The former is identified with the total electron density on the channel axis, and the latter with the average density of valence electrons in the solid. The quantity $\hbar k_c$ is the Bohm-Pines critical momentum transfer that determines the cutoff between individual and plasma excitations. It magnitude is on the order of $\hbar\omega_0/v_F$, where ω_0 and v_F are, respectively, the classical plasma frequency and Fermi velocity of the valence-electron gas. In general, the local and valence-electron-gas densities, NZ_{loc} and NZ_{val}, are not equal. Thus, in a sense the crystal is treated as an inhomogeneous electron gas. This model provides excellent agreement with the above referenced channeling data for MeV-energy protons penetrating through thin silicon crystals.

The inhomogeneous-electron-gas model is adapted to a treatment of the present problem in the following manner: First, a velocity-dependent effective charge $Z_i^*(v)e$ is introduced into Eq. (1) in place of the bare nuclear charge Z_ie. In this manner the slowing down of particles in *thick* targets can be treated, with a major aspect of the electron-capture processes taken into account. Second, Eq. (1) is amended by an explicit core-excitation term which takes into account the stopping-power contribution due to the excitation of inner-shell electrons. Such electrons cannot justifiably be treated in an electron-gas approximation, and their presence cannot be disregarded in light of the relatively high charge of the incident particles. Finally, the stopping-power parameters of Eq. (1) are evaluated for the cases of interest. These three aspects of the calculation are treated in the following paragraphs.

Effective Charge

Standard theoretical treatments applicable to amorphous media,[21,22] and to the free-electron gas,[23] indicate that stopping powers for energetic particles of charge Z_ie and velocity v take the form

$$-(dE/dx)\propto(Z_i^2e^2/v^2)L(v), \quad (2)$$

where $L(v)$ contains the dependence on the electronic properties of the medium. To the extent that $L(v)$ is dependent *only* on the target medium (as well as on v) it follows that a ratio of stopping powers

[19] M. Luntz, Ph.D. thesis, University of Connecticut, 1967 (unpublished).
[20] D. Pines, *Elementary Excitations in Solids* (W. A. Benjamin, Inc., New York, 1964).
[21] N. Bohr, Phil. Mag. **25**, 10 (1913).
[22] H. Bethe, Ann. Physik **5**, 325 (1930).
[23] J. Lindhard and A. Winther, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **34**, No. 4 (1964).

for two different particles, evaluated for a given medium and at a given velocity v, is entirely independent of the properties of the medium. The variation of such a ratio with velocity is then attributed to a variation in the charges of the incident particles. A velocity-dependent effective charge $Z_i^*(v)e$ can be defined as

$$(Z_i^*)^2=(-dE/dx)_i/(-dE/dx)_{\text{proton}}, \quad (3)$$

where the stopping powers are obtained at velocities high enough so that a proton is stripped of all electronic charge. Roll and Steigert[24] used this definition to generate effective-charge data from stopping-power measurements on a set of heavy ions incident on various solid and gaseous media. Typical data are displayed in Fig. 3, and are seen to be fit reasonably well by the function

$$Z_i^*=Z_i[1-\exp(-v/v_{\text{TF}})], \quad (4)$$

where v_{TF} is the Thomas-Fermi velocity of the incident particle, given by

$$v_{\text{TF}}=(c/137)Z_i^{2/3}. \quad (5)$$

This expression is used throughout the calculation to represent the charge of the incident particles.

Core Excitations

Tightly bound cole electrons are assumed to be excited by means of independent, two-body encounters, for which the corresponding energy transfers are obtained classically. The latter can be justified in terms of the highly localized nature of the incident-particle wave packets, and the rather well-defined character of the momentum transfers.[25] The classical

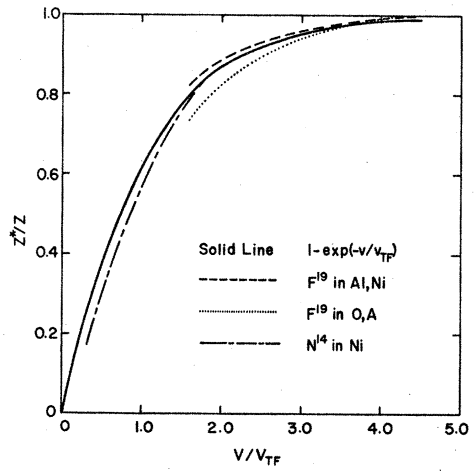

FIG. 3. Fractional effective charge for fluorine and nitrogen ions in various media as a function of the ratio of the ion velocity to the corresponding Thomas-Fermi velocity (Ref. 24). The solid curve represents the expression $[1-\exp(-v/v_{\text{TF}})]$.

[24] P. G. Roll and F. E. Steigert, Phys. Rev. **120**, 470 (1960).
[25] E. J. Williams, Rev. Mod. Phys. **17**, 217 (1945).

TABLE II. Energies of transitions to the lowest conduction band in NaI and CsI.

NaI Electron	ΔE (eV)	CsI Electron	ΔE (eV)
$I^-(5p)$	5.90	$I^-(5p)$	5.78
$I^-(5s)$	14.9	$Cs^+(5p)$	13.1
$Na^+(2p)$	33.3	$I^-(5s)$	14.8
$I^-(4d)$	56.4	$Cs^+(5s)$	24.9
$Na^+(2s)$	61.1	$I^-(4d)$	56.3
$I^-(4p)$	130	$Cs^+(4d)$	83.7
$I^-(4s)$	172	$I^-(4p)$	129
$I^-(3d)$	639	$Cs^+(4p)$	165
$Na^+(1s)$	1059	$I^-(4s)$	171
		$Cs^+(4s)$	211
		$I^-(3d)$	638
		$Cs^+(3d)$	747

stopping-power treatment of Bohr[21] proves readily adaptable to the case of core excitation for channeled particles, and leads to the expression

$$-\frac{dE^{\text{core}}}{dx}=\frac{4\pi(Z_i^*)^2e^4}{mv^2}N\sum_j c_j\ln\frac{(b_{\max})_j}{b}, \quad (6)$$

where c_j is the number of electrons in the jth electronic subshell of a target atom, $(b_{\max})_j$ is the maximum impact parameter for a particle-core-electron encounter, and the channel radius b plays the role of the minimum-impact parameter in Bohr's expression. The quantity $(b_{\max})_j$ is determined by an adiabaticity criterion described by Bohm,[26] and takes the form

$$(b_{\max})_j\propto(Z_i^*v)^{1/2}/\Delta E_j, \quad (7)$$

in which ΔE_j is the energy of transition from the jth subshell to the lowest unoccupied level. Upon inclusion of the above core-excitation term, the total stopping power becomes

$$-\frac{dE}{dx}=\frac{4\pi(Z_i^*)^2e^4}{mv^2}N\left(Z_{\text{loc}}\ln\frac{2mv}{\hbar k_c}+Z_{\text{val}}\ln\frac{k_c}{\omega_0/v}+\sum_j c_j\ln\frac{\beta(Z_i^*v/v_m)^{1/2}}{\Delta E_j}\right), \quad (8)$$

where β is a constant containing the channel radius b, and v_m is a reference velocity corresponding to $E/A=10$ MeV/amu (the maximum velocity considered in the present work). When the above expression is applied to the alkali-iodide channels, N is taken as the number of anion-cation pairs per unit volume, and the effective valencies Z_{loc} and Z_{val} are referenced to an ion pair.

The transition energies required for an evaluation of the core-excitation term are obtained by associating Hartree-Fock-Slater free-ion eigenvalues[27] with

[26] D. Bohm, *Quantum Theory* (Prentice-Hall, Inc., Englewood Cliffs, N.J., 1951).
[27] F. Herman and S. Skillman, *Atomic Structure Calculations* (Prentice-Hall Inc., Englewood Cliffs, N.J., 1963).

TABLE III. Electron-gas parameters.

Channel	N (10^{22} cm^{-3})	Z_{val}	ω_0(10^{16} sec^{-1})	k_c (10^8 cm^{-1})	Z_{loc}	S	I
NaI⟨100⟩	1.48	8	1.94	1.56	3.05	7.96	−6.00
NaI⟨110⟩					2.66	9.26	
CsI⟨111⟩	1.05	16	2.31	1.68	2.53	8.38	−11.0
CsI⟨100⟩					3.92	7.89	

the electronic shells of Na^+, Cs^+, and I^-, correcting for the Madelung energies of the NaI and CsI crystal structures,[28] and referencing the resulting crystal energies to appropriate conduction levels established by means of the empirical Hilsch-Pohl relation.[29] Implicit in this construction is the neglect of the alkali-iodide bandwidths in comparison with the interband gaps. This is a reasonable approximation, particularly when applied to the core electrons. Resulting transition energies for the valence shells and for the outer few core shells of NaI and CsI are listed in Table II.

Stopping-Power Parameters

To implement the stopping-power expression of Eq. (8) one must classify the electronic states of the solid into two categories: the tightly bound or core states, and the weakly bound or *valence-gas* states. Electrons in the latter states respond to an incident particle in a manner approximately characteristic of a free-electron gas. There are strong indications, of both an experimental[20] and theoretical[30] nature, that even for highly ionic crystals such states do indeed exist. However, their identification in NaI and CsI is at present a matter of conjecture. The outermost anion shells [$I^-(5p)$] of both crystals are immediately associated with the respective valence gases. For the identification of the remaining plasma states, appeal is made to rather simple considerations regarding the relative binding energies of the various electronic shells, as given in Table II, and their relative degrees of localization. It is concluded that the $I^-(5s)$ and $I^-(5p)$ shells comprise the valence gas in NaI (with $Z_{val}=8$), while these together with the $Cs^+(5s)$ and $Cs^+(5p)$ shells constitute the valence gas in CsI (with $Z_{val}=16$). All remaining states are regarded as core states.

Values for the remaining electron-gas parameters in Eq. (8), with the exception of NZ_{loc}, follow directly from the identifications made above. The local electron density for each channel is determined as follows: Thomas-Fermi-Dirac distributions for the electronic charge density of the crystal ions Na^+, Cs^+, and I^- are obtained,[31,32] and used to represent the ions comprising the rows bordering the channel. A direct superposition is made of the contributions of the various ions to the charge along the channel axis, and the resulting distribution is averaged along the axis to represent the local charge density. The use of the Thomas-Fermi-Dirac distribution at such a large radius is of doubtful validity, but no satisfactory alternative appears to be available.

The core-excitation term of Eq. (8) was computed as a function of v and was found to be given approximately by the linear expression

$$\sum_j c_j \ln \frac{\beta(Z_i^* v/v_m)^{1/2}}{\Delta E_j} \approx S\left(\frac{Z_i^* v}{v_m}\right)^{1/2} + I, \qquad (9)$$

where S and I are constants. This simpler form is used, with S and I obtained graphically.

Thus, in the foregoing manner all parameters required for the stopping-power calculation are obtained, and displayed in Table III. Several comments are in order. First, Z_{loc} averages about three electrons per ion-pair, or 1.5 electrons per ion, in contrast to the value of approximately four electrons per atom which was deduced previously[10] for the most open planar channels in silicon. This comparison seems quite reasonable in light of the relative *openness* of the alkali-iodide channels. Regarding the core-excitation parameters, one observes that the intercepts I are negative and have the same value for both channels in a given crystal. They serve to give the adiabatic cutoff for all core excitation, and are determined by the smallest excitation energy for the particular crystal. The slopes S are channel-dependent, and take on values that make the core-excitation least for the channels with the largest radii.

The calculated stopping-power curves for the NaI⟨100⟩ axis are displayed in Fig. 4, and are representative of the qualitative features exhibited by the four families of channel curves. A typical breakdown of a given channel stopping-power curve yields the following: Local excitation contributes ~30%,

[28] C. Kittel, *Introduction to Solid State Physics* (John Wiley & Sons, Inc., New York, 1967).
[29] R. S. Knox, in *Solid State Physics* edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1963), Suppl. 5.
[30] C. Horie, Progr. Theoret. Phys. (Kyoto) **21**, 113 (1958).
[31] H. Jensen, Z. Physik **101**, 141 (1936).
[32] P. Gombás, *Die Statistische Theorie Des Atoms Und Ihre Anwendungen* (Springer-Verlag, Vienna, 1949).

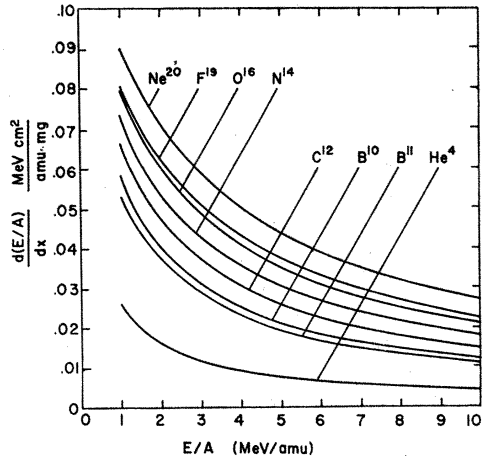

FIG. 4. Calculated values of the specific energy loss per nucleon as a function of the energy per nucleon for various heavy ions channeled along the NaI⟨100⟩ axis.

plasma excitation ~50%, and core excitation ~20%. This is only very approximate and is highly sensitive to the particular particle-channel combination considered and to the particle energy. A quantitative comparison among the various sets of curves demonstrates that in a given crystal the channel defined by the most dense rows of atoms corresponds to the least energy loss. In particular, the stopping power of the NaI⟨100⟩ axis is typically 3% lower than that of the NaI⟨110⟩ axis, while the CsI⟨111⟩ curves lie lower than those for the ⟨100⟩ axis by about 7%. This behavior is consistent with reported channeling data. Also, the stopping powers of the NaI channels lie typically 22% higher than those of the comparable channels in CsI. Higher stopping powers for NaI than for CsI also result from semiempirical calculations for randomly directed incident particles.[33] Finally, the channel stopping powers are found to range from approximately 20 to 25% of those characteristic of random incidence. This is seen for the case of CsI from a quantitative comparison between the calculated channel curves of Fig. 5 and the random curves of Fig. 6, where the latter were generated by scaling a semiempirical stopping-power relation for protons in CsI,[33] using the expression in Eq. (3). As was anticipated, the alkali-iodide channels exhibit a somewhat greater stopping-power reduction than do the channels in silicon or germanium.

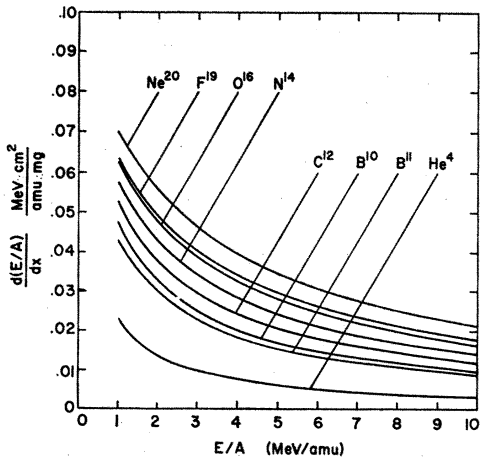

FIG. 5. Calculated values of the specific energy loss per nucleon as a function of the energy per nucleon for various heavy ions channeled along the CsI⟨111⟩ axis.

[33] R. B. Murray and A. Meyer, Phys. Rev. **122**, 815 (1961).

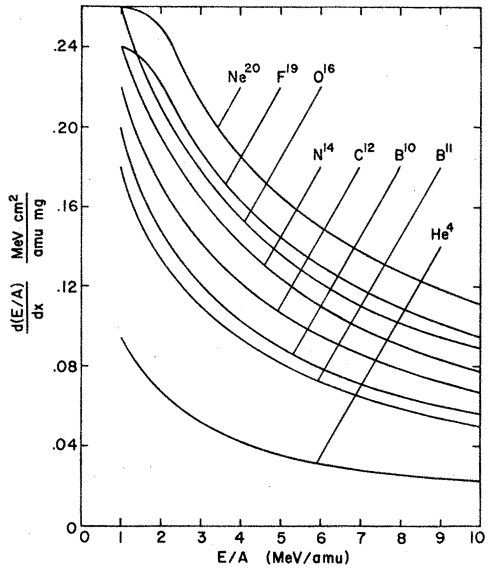

FIG. 6. Calculated values of the specific energy loss per nucleon as a function of the energy per nucleon for various heavy ions incident in a random direction in CsI (curves generated from the proton curve of Ref. 33).

While the results of the present calculation appear reasonable, a more definitive test awaits the availability of the appropriate channeling data. Range-energy relations generated from the calculated stopping-power curves by means of numerical integration facilitate a comparison with such data, in particular, a comparison with energy-loss measurements for particle transmission along the major symmetry axes, and through known thicknesses of crystal. The re-

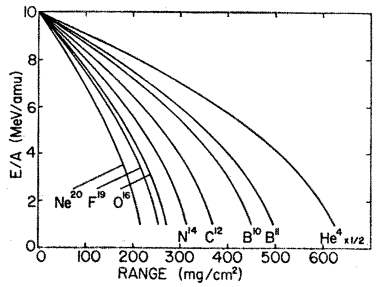

FIG. 7. Calculated range-energy relations for various heavy ions channeled along the NaI⟨100⟩ axis. Distances are measured from the point at which $E/A = 10$ MeV/amu.

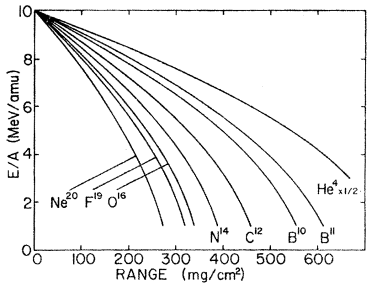

FIG. 8. Calculated range-energy relations for various heavy ions channeled along the CsI⟨111⟩ axis. Distances are measured from the point at which $E/A = 10$ MeV/amu.

sulting range-energy curves corresponding to the NaI⟨100⟩ and CsI⟨111⟩ axes are shown in Figs. 7 and 8, respectively. In performing such a comparison, knowledge of the sensitivity of the calculated stopping powers to deviations in the values of the various stopping-power parameters [see Eq. (8)] would be of value. Briefly, the parameter $Z_i{}^*e$ ranges (for the cases treated) from the bare nuclear charge Z_ie down to values typically $\sim 0.8Z_ie$, with a 10% deviation in $Z_i{}^*$ introducing approximately a 20% deviation in the stopping power. Similarly, a deviation as large as 50% in the value of Z_{loc} changes dE/dx by typically 15%. The sensitivity to Z_{val} must be related to the core-excitation term. In particular, an overestimate of Z_{val} results in a partially compensating underestimate of $(dE/dx)^{\text{core}}$. It is found for NaI that when *all* the excitable electrons are associated with the valence gas, the total stopping power is typically 50% higher than the values resulting from the more realistic choice of $Z_{\text{val}}=8$. This behavior indicates that valence-gas electrons are more responsive than core electrons, and consequently an overestimate of Z_{val} results in an overestimate of the total stopping power.

III. LUMINESCENT RESPONSE

Now that the required stopping-power relations have been established, the luminescence aspects of the problem may be considered. It is recalled that the Yale scintillation-efficiency curves (see Fig. 1) were constructed by means of a combination of luminescent-response data with the appropriate semiempirical stopping-power functions. To the extent that the original data (L versus E) represent truly *random* incidence, and to the extent that the semiempirical stopping-power curves approximate specific energy loss for truly *random* trajectories, the resulting scintillation-efficiency curves represent the scaling between incremental energy losses and corresponding light output for the given set of ions *randomly* incident on NaI(Tl) and CsI(Tl) scintillation counters. These relations are used, in conjunction with the stopping-power curves of the present study, in order to obtain a prediction of the luminescent response to the same set of ions incident along major axial channels. Implicit in this approach is the assumption that the dependence of dL/dE on dE/dx is the same for channeled particles as for randomly incident particles.

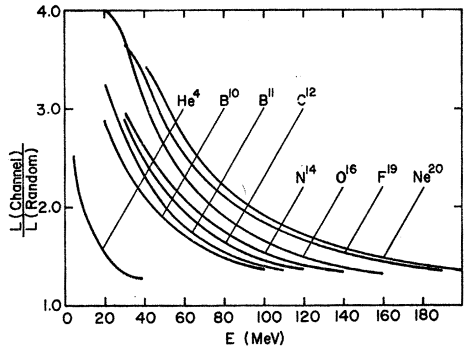

FIG. 9. Calculated ratio of the luminescent response of NaI(Tl) to channeled versus randomly incident heavy ions. The values of L(random) are obtained from Ref. 1.

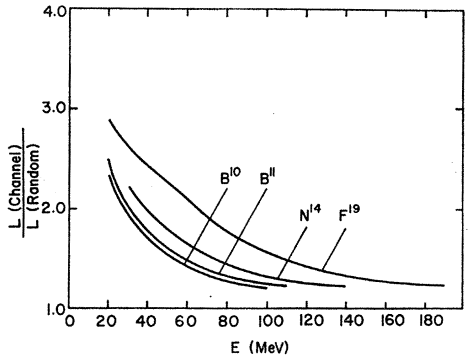

FIG. 10. Calculated ratio of the luminescent response of CsI(Tl) to channeled versus randomly incident heavy ions. The values of L(random) are obtained from Ref. 2.

A test of the validity of this assumption awaits a detailed theoretical analysis of scintillation efficiency for channeled particles.

Upon comparison of the various channel stopping-power curves with the corresponding scintillation-efficiency data, it is seen that for all cases treated, and over the entire range of interest, the channel stopping powers remain lower than the values at which the scintillation efficiency begins to fall. Thus it follows that for a given channeled particle the scintillation efficiency is constant, i.e., the luminescent response is linear and is given by the simple relation

$$L(\text{channel}) = (dL/dE)_{\max}E, \qquad (10)$$

where $(dL/dE)_{\max}$ is taken directly from the appropriate scintillation-efficiency curve in Fig. 1. In this manner one channel-response curve is generated for each crystal, and is assumed applicable to at least the two most prominent axial channels. From the manner in which the curves are obtained, it follows

that the channel response is automatically normalized to the Yale (random-response) data, and thus a comparison in the form of a ratio of L(channel) to L(random) is meaningful. Such a comparison is shown in Figs. 9 and 10 for NaI(Tl) and CsI(Tl), respectively. The resulting values correspond to the limiting case of channeling over an entire trajectory, and thus represent upper limits to the relative light output which a given channeled particle is capable of producing. These upper limits are seen to be fairly large for particles that have relatively low incident velocities. This is attributable to the random particle's experiencing a reduced scintillation efficiency over its entire trajectory, in consequence of its low velocity and therefore high specific energy loss. At high incident velocities the response ratio is about the same for all particles in a given crystal. In particular, channeled particles incident at $E/A=10$ MeV/amu in NaI(Tl) are capable of producing about 34% more light than are the corresponding random particles. In CsI(Tl) the increase in light output can reach 24%. Thus it is established that the luminescent response of NaI(Tl) and CsI(Tl) to energetic heavy ions is indeed anisotropic, with particle incidence parallel to major crystal axes corresponding to larger than normal light production.

IV. DISCUSSION

The only assumption about the process of luminescence required in Sec. III is that the dependence of dL/dE on dE/dx is the same for channeled as for randomly incident particles. Nevertheless, it is of interest to speculate on the mechanism of luminescence in order to gain more physical insight into the effects of channeling. The luminescent emission is characteristic of substitutional Tl^+ ions, to which the excitation is transferred. Murray and Meyer[33,34] assumed excitons were the dominant energy carriers, and attributed the reduction in dL/dE for high dE/dx to depletion of activators in the ground state. However, Gwin and Murray[35] showed subsequently that activator depletion is not significant, and more recent work[36–38] suggests that energy transfer is accomplished primarily by binary diffusion of electrons and holes which are successively trapped at Tl^+ sites. The hole exists as a V_k center, which diffuses by thermal reorientation.[39,40] Both Gwin and Murray[41] and Pooley[42] have shown that a process that competes with successive trapping of electrons and holes at impurity sites is the direct recombination of an electron and a hole to form a self-trapped exciton (an electron trapped at a V_k center).[43,44] At room temperature, the exciton subsequently decays nonradiatively,[45] and thus makes no contribution to the luminescence. For high ionization density (high dE/dx), this competing process should have a rate proportional to the product of electron and hole concentrations,[46] and could thus account for the decrease in dL/dE with increasing dE/dx. The enhanced scintillation efficiency for channeled particles may then be explained qualitatively by the reduced ionization density, which tends to favor radiative recombination of electron and holes by successive trapping at activator sites over nonradiative recombination at ordinary lattice sites.

Several avenues of further investigation that emanate from the present work are now suggested. First, while the anisotropy of the luminescent response has been accounted for, in particular, axes preferential for luminescence (crystal channels) have been identified, the corresponding acceptance angles calculated, and the luminescent response to channeled particles obtained and compared with the random response, it nevertheless follows, in light of the many assumptions made during the calculation, that the reliability of the theory is uncertain. Therefore, a detailed experimental analysis of the anisotropy is suggested. Such a study would appear to be well worth while, particularly since the theory indicates a fairly pronounced effect.

An anisotropic scintillation response associated with channeling has been observed[47–49] in single crystals of anthracene. However, contrary to the conclusions of the present paper, there is a *dip* in the scintillation efficiency for channeled particles in thick anthracene crystals. Although the energy-transfer mechanism is different in anthracene and alkali iodides, nevertheless dL/dE decreases with increasing dE/dx in both cases. Thus the assumption that the dependence of dL/dE on dE/dx is the same for channeled as for randomly incident particles does not appear to be valid for anthracene. This assumption must be questioned for alkali iodides as well. The distribution of energy loss among local, plasma, and core excitations is substantially different for channeled and randomly incident particles, and these three excitation processes may not be equally efficient in ultimately producing ionization density. Thus there is a need for further theoretical investigation of the mechanism of energy transfer to the activators.

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Additional areas of investigation are indicated by the requirements for a critical analysis of the various features of the present stopping-power model. Since a correlation with energy-loss measurements for channeled particles in thin NaI and CsI single crystals would provide the most direct test of the over-all model, it follows that such data would be highly desirable. Plasma-excitation measurements in NaI and CsI would provide useful information as to the identity of the respective sets of valence-gas electrons, and a measure of the extent to which the plasma electrons are capable of exhibiting free-electron-gas-like behavior. Finally, an independent experimental determination of the density of residual charge down the various axial channels would also be of value.

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Mean Excitation Potential of Light Compounds*

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The mean excitation potentials I of Al, Si, Al_2O_3, SiO_2, and $C_6H_8O_2$ were determined to an accuracy of $\pm 0.5\%$ by fitting theoretical range functions to experimental energy-loss data. The I values for the compounds were found to be 7 to 11% larger than the corresponding values calculated for mixtures using Bragg's rule.

1. INTRODUCTION

IT was the purpose of this study to measure and compare the mean excitation potentials I of light compounds with those of their constituent elements. The quantity I appears in the well-known formula for the stopping power $\langle dT/dx \rangle = S(T)$ for heavy charged particles in homogeneous absorbers.[1,2]

$$S(T) = (0.30706/\beta^2)(Z/A)\rho\{\ln[2mc^2\beta^2/(1-\beta^2)] - \beta^2 - \ln I - \sum_i C_i/Z\}, \quad (1)$$

where

T = energy of the incident particle,
$\beta = v/c$ = particle velocity,
Z = charge number of absorber,
A = atomic or molecular weight of absorber,
mc^2 = electron rest energy = 511 006 eV,
C_i = shell correction for shell i of the absorber atoms,
ρ = density of absorber material.

The mean excitation potential is defined as[1]

$$\ln I = \sum_i f_i \ln E_i,$$

where f_i is the optical dipole oscillator strength for excitation of an absorber atom from its ground state to the excited state i of energy E_i. The sum extends over all excited states of the atom. The theoretical calculation of I has proved to be a formidable task and has so far only been attempted for very simple atoms.[3] However, a method of determining I experimentally is discussed below.

The shell corrections C_i in Eq. (1) have been calculated for the K shell[4] and the L shell,[5] assuming hydrogenic electron wave functions. They appear to fit experimental data quite well.[2] Higher shell corrections however, are still only little known.[2] We have therefore confined this study of I to Si, Al, and lighter atoms where the higher shell corrections may be neglected.

We now establish the formula for the stopping power in absorbers consisting of a mixture of atomic elements (not a compound), assuming no interaction between

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