The Scintillation Response from NaI(Tl) Crystals Under Bombardment with Positive Ions of Energies 60–600 kev

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The scintillations of NaI(Tl) under ion bombardment have been investigated, using monoenergetic H, D, He, and Ne ions in the kinetic energy range 60–600 kev. The light output caused electrical pulses in a photomultiplier tube (Type EMI 5311), which were then sorted in magnitude by means of a single channel pulse height analyzer. Single peaked pulse-height distributions were observed for each ion energy. The most probable pulse height was found to increase linearly with energy for each of the ions. The pulse height (most probable) *versus* energy lines for H, D, and He were found to extrapolate through the origin, with an experimental uncertainty of perhaps 10 kev in the energy intercept. The slopes of these lines, giving the relative scintillation efficiencies, were observed to be in the ratios $1:0.96\pm0.03:0.75\pm0.05:0.54\pm0.03$ for the H, D, He, and Ne ions, respectively. It is concluded that the response of NaI(Tl) to protons is approximately the same as for electrons of the same energy, since an extrapolation of the proton pulse-height curve to 661 kev gave the same pulse height as was observed for the photopeak of the Cs^{137} gamma-ray of that energy. The importance of "hard" (i.e., nuclear) collisions, which may not produce scintillations, is brought forward as a factor which may enter into the explanation of the lowered scintillation efficiency for the heavier ions.

INTRODUCTION

THE introduction of scintillating crystals with photomultiplier tubes to pick up their light pulses marked an important step forward in the detection and analysis of gamma-rays because of the high detection efficiency, much greater than that of the ionization chamber at ordinary pressures, or the Geiger counter. Thus there have been many investigations of the response of such scintillators to gamma-rays.

In the detection of heavy positive ions the crystal detector does not necessarily have the advantage of greater efficiency in the sense of counting a larger fraction of the particles incident on it, but has other advantages such as simplicity and reliability. The visual observation of scintillations, such as those of activated ZnS crystals under alpha-particle bombardment, has been known for many years, but it was the relatively recent development of photomultiplier tubes giving a sensitive, objective, and quantitative electrical response to the light pulses of the scintillator which has stimulated the current interest in this method of detection.

Several investigators have measured the pulse height (i.e., peak of the pulse-height distribution curve) *versus* particle energy using natural alpha-particles which were varied in energy by being slowed down in various path lengths of air.[1] Because of straggling effects and uncertainties in the range-energy relation, the measurements could not be extended with any great accuracy below kinetic energies of 1 Mev. In the years 1948–1951 a number of such papers were published, but only three of them (Johansson, Michaelis, and Lovberg) employed NaI(Tl) as the scintillator. These investigations showed that the organic scintillators as a group give a nonlinear response to energy expended when bombarded with alpha-particles, but for inorganic crystals the light is more nearly directly proportional to the particle energy. In 1951 the work of Frey *et al.*[2] using protons in the energy range 0.4 Mev to 3.7 Mev showed that anthracene, for instance, has a nonlinear response.

The recent work of the Princeton investigators[3] in bombarding NaI(Tl) with protons of energies between 5 and 18 Mev has indicated a strictly linear response, which furthermore extrapolated to the origin (zero pulse height at zero energy) within the limit of their experimental error. The University of Illinois group,[4] bombarding NaI(Tl) with protons having energies above 3 Mev, and with deuterons and alpha-particles of similar energies, observed a linear response for protons and deuterons, but detected deviations from linearity for the helium ions.

In seeking a suitable particle detector for use in connection with a new spherical electrostatic analyzer the authors concluded that NaI(Tl) might be useful for the

TABLE I. Constants of the spherical analyzer.

r_2	radius of outer sphere	25.5806 ± 0.0003 cm
r_1	radius of inner sphere	25.1628 ± 0.0003 cm
δr	gap width of analyzer	0.4178 ± 0.0005 cm
b_2	outer radius of beam limiting annulus	0.5159 ± 0.001 cm
b_1	inner radius of beam limiting annulus	0.4167 ± 0.001 cm
w_t	projection of width of target annulus on sphere diameter	0.0992 ± 0.002 cm
w_d	width of annular opening at detector	$0.10\ \pm0.01$ cm

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[1] J. Broser and H. Kallmann, Ann. Physik. **3**, 317 (1948); S. A. E. Johansson, Arkiv. fys. **2**, 171 (1950); E. G. Michaelis, Helv. Phys. Acta **23**, 155 (1950); J. B. Birks, Proc. Phys. Soc. (London) **63**, 1294 (1950); Harding, Flowers, and Eppstein, Nature **163**, 990 (1949); B. Collinge and E. J. Rolbins, Nature **166**, 1109 (1950); R. H. Lovberg, Phys. Rev. **84**, 852 (1951).

[2] Frey, Grim, Preston, and Gray, Phys. Rev. **82**, 372 (1951).

[3] Franzen, Peelle, and Scherr, Phys. Rev. **79**, 742 (1950); J. G. Likely and W. Franzen, Phys. Rev. **87**, 666 (1952).

[4] Taylor, Remley, Jentschke, and Kruger, Phys. Rev. **83**, 169 (1951); Jentschke, Eby, Taylor, Remley, and Kruger, Phys. Rev. **83**, 170 (1951); Taylor, Jentschke, Remley, Eby, and Kruger, Phys. Rev. **84**, 1034 (1951).

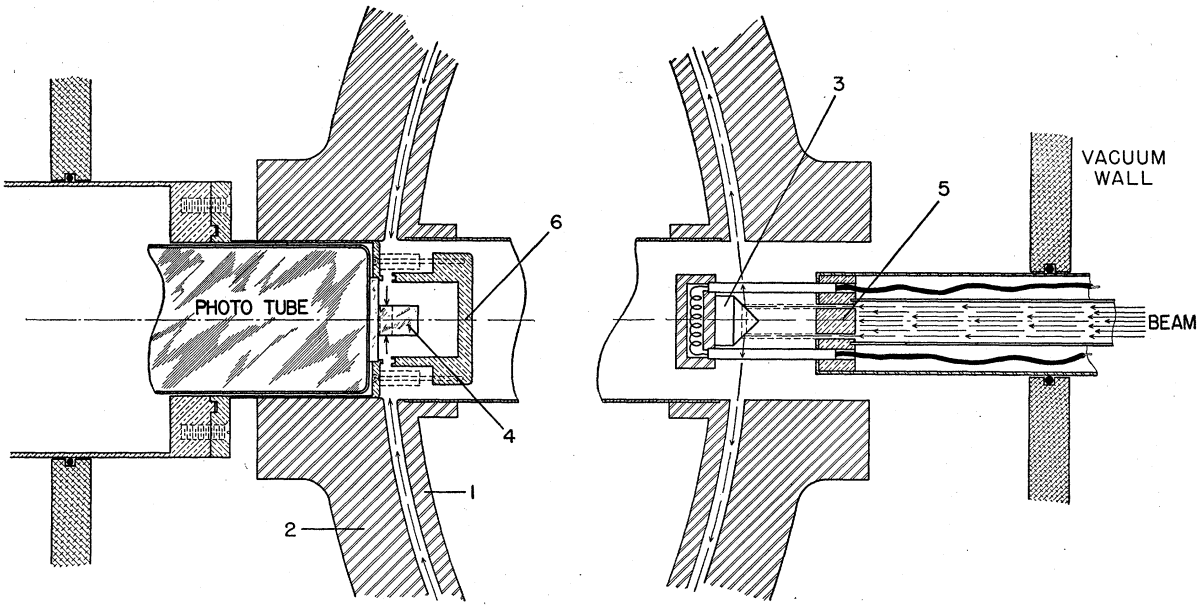

FIG. 1. Schematic diagram of the spherical electrostatic analyzer showing the scattering target and the detector crystal.

detection of low energy (down to 50 kev) particles in view of the relatively large light output and the indications of linear response to energy expended. The hygroscopic property of the material is a disadvantage, but the construction of an air-tight housing, which can be closed in a dry-box after the crystal has been mounted in place, and then opened in vacuum for bombardment purposes, has enabled the crystals used in this work to be maintained without deterioration indefinitely.

The present paper gives the results of bombarding two NaI(Tl) crystals with monoenergetic ions focused on their surfaces by the electrostatic analyzer.

II. APPARATUS

The equipment used in this work may be divided into the following categories: (A) the low voltage arc ion source; (B) the Cockcroft-Walton accelerator, with magnetic deflection through 15° used to select the type of ions desired; (C) the beam stop and the thick target which scattered the ion beam, producing a continuum of energies; (D) the electrostatic analyzer which selected from the continuum a small number of relatively monoenergetic ions and focused them on the detector; and (E), the NaI(Tl) scintillation detector and its associated electronic circuits.

A. Ion Source

The ion source was of the capillary arc type with a perforated probe using accelerating voltages of 1–5 kv to extract the ions.[5] The source operated satisfactorily on the various gases used, namely H_2, D_2, He, and Ne. Changing from one gas to another took perhaps fifteen minutes, and thus several ion types could be compared during a run. As each new ion type was tested for its scintillation response, it was at once compared with responses from protons. An appreciable number of doubly charged neon ions were found in the beam when neon was fed into the low voltage arc ion source. These Ne^{++} ions could thus be accelerated up to 1-Mev kinetic energy in the accelerator.

[5] S. K. Allison, Rev. Sci. Instr. **19**, 291 (1948).

B. Cockcroft-Walton Accelerator

The Cockcroft-Walton circuit was of the voltage sextuplet type operating in the range 0–500 kev. Either a 60-cycle or 540-cycle input supply voltage could be used.

C. Beam Stop and Target

Figure 1 shows, somewhat schematically, the arrangement for bombarding NaI(Tl) crystals. The conical target (3) and the cylindrical detector crystal (4) were mounted at opposite ends of a diametral shaft which extended through the analyzer and were coaxial with the direction of the incoming ion beam. The beam stop (5) served as an annular slit to define the primary beam before scattering. After passing through the beam stop, the beam particles about to impinge on the target were moving longitudinally in a cylindrical shell of approximately (see Table I) 0.5-cm mean radius and 0.1-cm thickness. The angle of the target cone was 90 degrees. Scattered particles, degraded in energy, left the target and those emitted in the proper range of angles (mean angle = 85°16′ with respect to the beam axis) and with energy appropriate to the charge of the analyzer, were focused upon the crystal detector. Both thick gold and thick beryllium targets were used during the course of the work. A beryllium target was used in

the experiments in which the disintegration deuterons from the $Be^9(p,d)$ reaction were focused on the crystal.

The scattering of the beam produced a continuous distribution in energy of the scattered particles, the energies lying below an upper limit, E_{max}, which is approximately

$$E_{max}=E_0(M-m)/(M+m),$$

where M and m are the masses of the target and projectile nuclei, respectively, and E_0 is the beam energy.

Although the presence of doubly charged scattered ions could be detected, the analyzer was always operated in a region in which only the singly charged ions would be focused.

When neon was used in the ion source, an appreciable number of ions were accelerated in the doubly charged state and could be magnetically deflected onto the target. After scattering from the target, however, these neon ions were found to be mostly in the singly charged state and were focused by the analyzer as such. The upper energy limit for neon ions scattered at 85° from a gold target is approximately 80 percent of the beam energy before scattering. This provided a means of studying the scintillation response of neon ions of energy greater by a factor of about 1.6 than the energy attainable for protons.

D. The Spherical Electrostatic Analyzer

The deflecting electrostatic field of the analyzer was located in a spherical shell between the outside surface of an inner, hollow metallic sphere (1) and the inside surface of two outer covers in the form of hemispherical shells (2). The focal properties of such an analyzer have been calculated by Purcell[6] and others.

The analyzer was always operated with the outer covers at ground potential and the inner sphere charged negatively. If we neglect the fringing electric field at the 5.08 cm diameter apertures where the diametral shaft pierced the spherical surfaces, we may write

$$W=kze|V|=[r_1r_2/(r_2^2-r_1^2)-\tfrac{1}{2}]ze|V|,$$

for this type of charging of the analyzer. W is the energy in kev of the particle of charge ze selected by the analyzer when the potential in kev of the inner spherical surface is $-|V|$. r_1 and r_2 are the radii of the inner and outer spheres, respectively. From the dimensions given in Table I, we obtain

$$k=29.861\pm0.043.$$

The homogeneity in energy of the scattered beam after passing through the detector slit can be estimated as follows. The maximum variation in orbit diameter is 0.2 cm when the effective target and detector slit widths are each equal to 0.1 cm. The orbits in the inverse square law field between the spheres are ellipses, and the energy content of a Keplerian elliptic orbit is proportional to its major axis. Thus the maximum spread in energy is

$$\Delta W/W=0.2/(2\times25.37)=0.39 \text{ percent}.$$

E. The NaI(Tl) Crystal Detector and Associated Apparatus

Two crystals of NaI(Tl) were used. They were cylindrical in form, approximately 1.27 cm in diameter and 1.27 cm high. The photomultiplier tube, an EMI 5311 tube, was enclosed in a brass housing and inserted through the vacuum wall of the analyzer into the diametral shaft. The end of the brass housing carried a quartz window, against which the crystal was pressed by a spring. The crystal was separated from the spring by a brass cap which was lined with shiny aluminum foil to improve the light collection. The presence of the reflector produced an increase of almost a factor of 2 in the pulse heights. Optical contact between crystal and quartz and between quartz and photomultiplier tube was improved by films of silicone grease.

The large brass cap (6) surrounding the crystal, reflector, and spring assembly, served two purposes. (a) The cap could be raised without breaking the vacuum in the analyzer by removing the photomultiplier tube from its housing and, with the aid of two special screwdrivers, rotating the two screws indicated in the drawing. These screws threaded into the cap, and by means of them it could be rasied or lowered, thus changing the width of the annular aperture through which the particles passed before striking the crystal. The cap thus served as the movable part of a slit which, together with the annular beam aperture previously described, determined the resolving power of the analyzer. Furthermore, if the crystal had to be removed from the analyzer, the cap could be pulled down against a rubber gasket in the photomultiplier housing. Thus the crystal, which is hygroscopic, could be protected from the moisture in the air while the assembly was being carried to and from the dry box.

The two cylindrical crystals of NaI(Tl) used in this work were purchased at different times (approximately two years apart) from the Harshaw Chemical Company. It was expected that if there were serious differences between the specimens, such as different thallium content, this would become evident in the relative response to ion bombardment.

Since the heavy ions we used are stopped very near the surface of the crystal (i.e., within 10^{-4} cm) it was important that the surface be cleaned and kept in good condition. Chemical methods were avoided in preparing the surfaces for ion bombardment. The crystals were polished in a dry box, with various grades of emery paper used in the early stages, and the final stages of polishing carried out with felt. A small motor-driven turn-table was installed in the dry box and either

[6] E. M. Purcell, Phys. Rev. 54, 818 (1938); Browne, Craig, and Williamson, Rev. Sci. Instr. 22, 952 (1951).

allowed the crystal to be rotated while the brasive was applied to the cylindrical surface, or permitted the emery paper or felt to be rotated while the crystal was held fixed.

The electronic circuit into which the pulses from the photomultiplier tube were fed has been briefly discussed elsewhere.[7] The photomultiplier output was fed into a preamplifier, which in turn fed into a Los Alamos Model 500 linear amplifier. The pulses from the amplifier were analyzed using a single channel discriminator.

RESULTS

By repeated determination of the pulse-height distributions for various proton energies, it was ascertained that the proton pulse height *versus* energy curve for each NaI(Tl) crystal did not deviate significantly from a straight line extrapolating to the origin. The curve for protons was then chosen as the standard, and a curve for protons was also taken whenever a run was made using one of the other ions, D, He, or Ne. In some cases, proton points were taken both before and after a run on another ion. In this way, it was possible to compare directly the scintillation response of each ion with that of protons, and to minimize possible errors due to drifts in the electronic equipment.

The results of a typical run using protons and deuterons on NaI(Tl) crystal No. 2 are plotted in Fig. 2. The results of typical runs using helium ions and neons ions are also shown. The latter curves were normalized to bring all the corresponding proton slopes to the same value. The normalization factor in each case differed from unity by less than 10 percent.

The curves were all found to be linear in the regions investigated. Within the limits of error, the proton, deuteron, and helium ion curves extrapolated through the origin, having an energy intercept of less than 10 kev. For neon, however, a positive energy intercept was observed which was different for the two crystals. It was about 30 kev for NaI crystal No. 2 and about 70 kev for No. 1.

Table II gives the average relative values of the slopes, referred to as values of dL/dE, observed on crystals No. 1 and No. 2. The proton slope has been assigned the value unity.

For comparison with light pulses caused by electrons liberated inside the crystal we measured the pulse-height distribution caused by the absorption of the 661-kev gamma-ray[8] of Cs^{137}. In order to make the comparison meaningful, it was necessary to use the same geometrical and optical arrangement as for counting particles. The photomultiplier tube was slipped out of its housing and replaced after a thin source of Cs^{137} had been taped to its front face. The source was attached to the periphery of the tube where the light path from the crystal into the photomultiplier was not affected. Pulses from the complete absorption of the 661-kev gamma-ray in the crystal were compared with those from protons striking its surface without disturbing the arrangement and during the same run.

The geometry was however not well suited to the measurement of gamma-rays. The crystal was too small and the presence of the heavy metal walls of the spherical analyzer near the crystal caused it to be bombarded with many secondary electrons degraded in energy below the maximum of 661 kev. Thus the photopeak was not well separated from a continuous distribution of pulses on its low energy side. The high energy side of the peak was sharp, however, and its position could be located to about 10 percent in pulse height, as is plotted in Fig. 2.

The fact that the rather inaccurate point from Cs^{137}

TABLE II. Relative values of the slope (dL/dE) of the pulse height *versus* energy curve.

Incident particle	Energy range investigated	$(dL/dE)/(dL/dE)_H$ Crystal No. 1	Crystal No. 2
H	60–400 kev	1.00	1.00
D	60–630	0.94	0.98
He	80–360	0.70	0.79
Ne	180–525	0.53	0.55

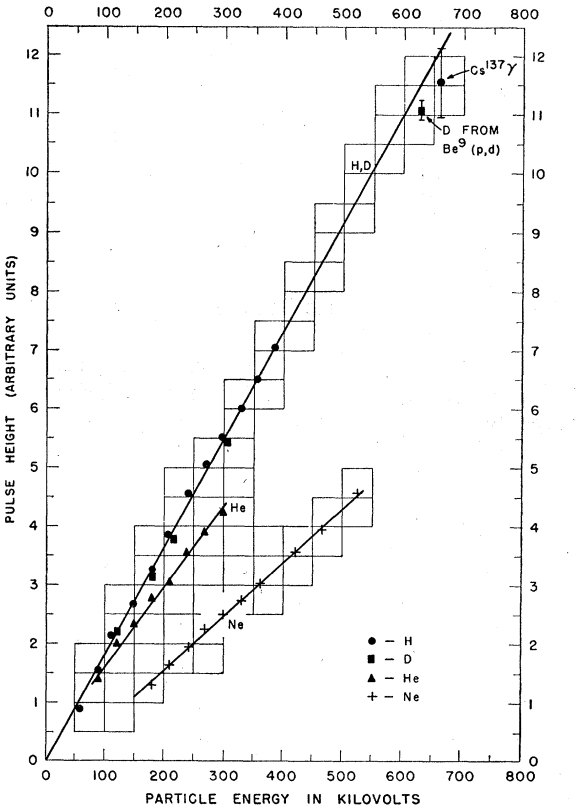

FIG. 2. Observed scintillation pulse heights from NaI(Tl) as a function of ion energy.

[7] H. Casson, Phys. Rev. **89**, 809 (1953).

[8] L. M. Langer and R. D. Moffat, Phys. Rev. **78**, 74 (1950).

falls, within its limits of error, on the extended proton curve indicates that electrons and protons are approximately equal in efficiency as measured by the output of light per unit of energy expended in the crystal.

Resolving Power of the Crystal Detector

As is well known, the energy resolution attainable in a crystal spectrometer depends on the particular photomultiplier tube used and may be different for different crystal specimens. Figure 3 shows the resolution, which is defined as the full width at half-maximum of the pulse-height distribution from monoenergetic ions, expressed as the percentage of the pulse height associated with the peak of the distribution. Energy resolutions applicable to helium ions and protons are shown. In the range of our investigations the resolution varies roughly as $E^{-0.7}$, but this result is probably not of general significance.

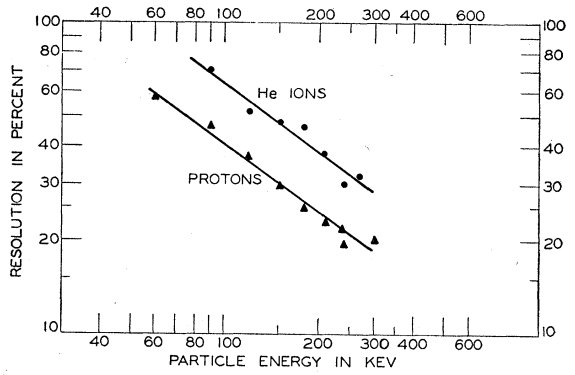

FIG. 3. Resolving power of the crystal-photomultiplier combination.

DISCUSSION

A quantitative explanation of the linearity of the curves of light output *versus* energy, and of the variations of dL/dE for the different particles cannot be given at present. However, we may make the following comments. In the first place, the fact that the proton, deuteron, and helium ion curves extrapolate to the origin indicates that there is not a "dead" or nonscintillating layer at the crystal surface.

Furthermore, we see that if a certain amount of energy dE is liberated in the crystal from a moving proton, there is approximately twice as much response from the photomultiplier tube as is the case when the same amount of energy is abstracted from a moving neon ion. Various hypothetical explanations at once suggest themselves.

A. The specific ionization from the heavier ions is great enough to partially saturate the ability of the crystal to respond, hence the efficiency of response decreases.

B. The spectrum of the light excited by the heavier ions differs from that of electrons and protons, and the energy is shifted to a region in which the photomultiplier tube is relatively insensitive.

C. In the case of the heavier ions the direct transfer of momentum to the atoms of the crystal becomes of importance in the degradation of ion energy and does not result in the excitation of scintillations.

It seems demonstrable that hypothesis A, alone, is not capable of explaining the observations. Our laboratory is now measuring the stopping power of gases for various ions, including helium and neon ions, and we will anticipate here some of the results which will be published in detail elsewhere. The stopping powers in gases were measured in a beam geometry such that if an ion suffered a deflection of angle greater than about 5×10^{-4} in the gas, it would not pass through the analyzing slits, and hence its energy loss would not contribute to the measured result. This affects the interpretation of the results on dE/dx for neon ions, and will be discussed later. The data on the stopping power of gold for helium ions, which are included with the gas data in Table III, were recalculated from the results published by Wilcox.[9] Although Wilcox's values as originally published for helium ions in gold are now known to be in need of a correction factor, we are fortunately able to compute that factor, because Wilcox published the stopping power of this same foil for protons. Using more recent values for protons in gold,[10] we are able to compute the effective thickness of the gold foil from the proton data. The correction thus indicated for Wilcox's gold data has increased his dE/dx values about 14 percent.

Taking the values of dE/dx for helium ions between 100 and 400 kev in argon and in gold as indicative of the variation that would obtain in NaI(Tl), it is found that in this energy range the values of dE/dx for helium ions in the crystal will probably change by a factor between 1.5 and 2. The approximate linearity of the light pulse response curve from helium ions throughout this region of energy indicates that these relatively large changes in dE/dx are not followed by commensurate changes in dL/dE, and hence if saturation effects are operating, we are in a relatively insensitive portion of the saturation curve.

On the other hand, turning to the results for neon ions, we find that the values of dE/dx for them will not be much different from the values for helium ions of the same energy. The geometry in which the measurements in the gases were made is such that only energy transfers to individual electrons, resulting in electron ionization, excitation, or exchange contributed to the measured stopping power (see the following discussion of hypothesis C), and the results show that the losses due to this type of interaction were approximately 15 percent greater in neon than in helium ions of the same energy. The scintillation efficiencies for the two ion types are

[9] H. A. Wilcox, Phys. Rev. **74**, 1743 (1948).
[10] S. D. Warshaw, Phys. Rev. **76**, 1759 (1949); D. Kahn, Phys. Rev. **90**, 503 (1953).

TABLE III. Atomic stopping powers due to the summation of events which have deflected the particles through angles less than 10^{-3} radian in argon and in gold. Units: $\{(\text{ev}\times\text{cm}^2)/\text{atom}\}\times 10^{15}$.

Kinetic erngy in kev	H in A (gas)	He in A (gas)	He in Au (metal)	Ne in A (gas)
50	33	...	...	...
100	32	...	52.3	...
150	28	53.0	63.8	...
200	25	59.5	71.9	62.1
250	22	65.5	79.1	69.5
300	20	70.5	86.0	75.8
350	18	75.0	91.9	81.7
400	16	79.3	97.1	87.0

however markedly different; $(dL/dE)_{\text{Ne}}=0.7(dL/dE)_{\text{He}}$. Thus, although this semiquantitative argument by no means excludes the possibility of saturation effects in scintillation, it indicates strongly that other factors must enter.

Hypothesis B is capable of a straightforward spectroscopic test, but we have not carried out such an investigation.

Rough calculations concerning hypothesis C can be made using a formula due to Bohr,[11] which estimates the loss of energy due to "hard" collisions, and was evolved in the attempt to interpret the low energy end of the fission fragment range-energy curve. This formula is

$$\left.\frac{dE}{dx}\right|_{\text{hard}}=\frac{4\pi e^4 Z_1^2 Z_2^2}{M_2 V^2}\log\left[\frac{M_1 M_2}{M_1+M_2}V^2\frac{a_{12}^{\text{scr}}}{Z_1 Z_2 e^2}\right]\frac{\text{erg cm}^2}{\text{atom}},$$

where Z_1, Z_2 are the atomic numbers of the moving particle and of the stopping nucleus respectively; M_1, M_2 are respectively the atomic masses of particle and nucleus; V is the velocity of the particle in cm/sec, and a_{12}^{scr} is the distance between particle and nucleus when the electronic screening has set a limit to the action of the nuclear charges. The quantity a_{12}^{scr} may be calculated with sufficient accuracy from

$$a_{12}^{\text{scr}}=h^2[(1/Z_1)+(1/Z_2)]/(4\pi^2 me^2),$$

in which m is the electronic mass.

Using collisions in argon as examples of collisions in a medium of atomic weight intermediate between sodium and iodine, the values listed in Table IV have been computed. The computed values for "hard" collisions of neon in argon show that in the total stopping process collisions of this type play an appreciable role. However, Bohr,[12] in his discussion of this matter, has shown that the "hard" scattering which results in angular deflections less than an angle θ_a' can be neglected in its contribution to dE/dx. The angle in question is given by

$$\theta_a'=s_{\min}/a_{12}^{\text{scr}},$$

where $s_{\min}$ is the closest approach of the two nuclei in a head-on collision and is given by

$$s_{\min}=(2Z_1Z_2e^2)/\mu V^2,$$

where μ is the reduced mass of the two bodies. For neon ions of 400-kev kinetic energy in argon we compute that $\theta_a'=6.3\times 10^{-2}$. Since an angular deflection greater than 5×10^{-4} would throw the ion out of our detecting system, contributions to our gas absorption measurements from losses in hard collisions were negligible, and we must ascribe the measured dE/dx values to that part of the total stopping arising from electronic encounters.

TABLE IV. Estimate of the ratio of energy loss in hard collisions to that in electronic collisions in argon gas (estimates at 400-kev kinetic energy).

Moving particle	a_{12}^{scr} $\times 10^9$ cm	Computed dE/dx for "hard" collisions (ev cm²)/atom $\times 10^{15}$	Observed dE/dx from electronic encounters	Ratio of "hard" to electronic losses
H	5.6	0.02	16	0.12
D	5.6	0.04	25	0.2
He	2.9	0.25	79	0.3
Ne	0.82	15.0	87	17.2

For moving ions as heavy as neon, Table IV indicates that in our energy range the stopping by hard collisions in the crystal constitutes a considerable portion of the total energy loss; due to the high atomic number and mass of iodine the fractions will be considerably greater than the estimates given here for argon, which is chosen merely as a moderately heavy stopping medium in which some experimental data on neon ions is now available. If such collisions are ineffective in causing scintillations, they may play a considerable role in the observed lowering of the scintillation efficiency for heavy ions.

[11] N. Bohr, Phys. Rev. **59**, 270 (1941).

[12] N. Bohr, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **18**, 8 (1948), p. 20, Eq. (1.4.3); and p. 47, Eq. (2.3.6.)