

## Response of Some Scintillation Crystals to Charged Particles\*†

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(Received June 11, 1951)

The dependence of the response of anthracene, stilbene, and sodium iodide crystals in a scintillation detector on the type and energy of charged particles incident on the crystal has been investigated. Electrons with energies from 500 ev to 624 kev, deuterons and molecular hydrogen ions with energies of 1 to 11 Mev, protons of 1 to 5 Mev, and alpha-particles of 4 to 21 Mev were employed. With the exception of protons and deuterons in sodium iodide, which gave a linear response over the entire energy region investigated, plots of pulse height vs energy for heavy particles gave a nonlinear relation for low energies tending toward linearity with increasing particle energy. Electrons in anthracene and stilbene show a nonlinear response below 100 kev, while the sodium iodide curve is linear above 1000 ev. The response of anthracene, whose behavior may be typical of organic crystals, has been compared for different ionizing radiations by considering the variation of the specific energy loss. Both electrons and heavy particles seem to show regions of linear response which start at quite different values of  $dE/dx$ . As the specific energy loss increases the response becomes nonlinear, with the ultimate saturation of the specific fluorescence for large values of  $dE/dx$ .

### I. INTRODUCTION

THE fluorescence of certain crystals under bombardment by energetic particles has been known for some time. The combination of such a crystal and a photomultiplier tube results in a device suitable for electronic counting of individual particles entering the crystal.<sup>1</sup> A recent survey of the important properties of these crystals has been given by Hofstadter.<sup>2</sup> The greater part of the work done so far has been concerned with the response of various crystals to gamma-rays and high energy beta-particles, and Pringle<sup>3</sup> has given an excellent summary of the developments in this field. A relatively small amount of information has been reported con-

cerning the response of scintillation crystals to heavy particles,<sup>4-11</sup> but knowledge of this kind is essential to the successful employment of the scintillation counter technique in the field of quantitative energy measurements of heavy particles. The response of organic crystals to fast protons is particularly interesting due to the possibility of using such crystals for fast neutron energy measurements.

Protons of 1 to 5 Mev, deuterons and molecular hydrogen ions of 1 to 11 Mev, alpha-particles of 4 to 21 Mev from the University of Illinois cyclotron, and polonium alpha-particles were used in the measurements with heavy particles. The response of anthracene, stilbene, and thallium activated sodium iodide to these particles has been studied.

The investigation was then extended to a study of the fluorescence of these crystals with incident monoenergetic electrons. This extension was particularly directed toward an investigation of the fluorescence

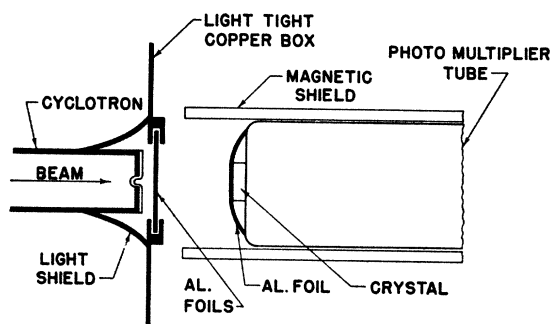


FIG. 1. Experimental arrangement for the measurement of the fluorescence of crystals to heavy particles. Note that the distance from the Nylon foil to absorber and from absorber to crystal are not to scale.

\* Assisted by the joint program of the ONR and AEC.

† Parts of this work were presented by one of us (C.J.T.) in partial fulfillment of the requirements for the Ph.D. at the University of Illinois.

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<sup>1</sup> H. Kallman, *Natur und Technik* (July 1947), M. Deutsch, Massachusetts Institute of Technology Technical Report No. 3 (December 1947); Marshall, Colman, and Bennett, *Rev. Sci. Instr.* **19**, 744 (1948); R. J. Moon, *Phys. Rev.* **73**, 1210 (1948); P. R. Bell, *Phys. Rev.* **73**, 1405 (1948); R. Hofstadter, *Phys. Rev.* **74**, 100 (1948); H. Kallmann, *Phys. Rev.* **75**, 623 (1949). W. Hanle, *Naturwiss.* **38**, 176 (1951).

<sup>2</sup> R. Hofstadter, *Nucleonics* **6**, No. 5, 70 (1950).

<sup>3</sup> R. W. Pringle, *Nature* **166**, 11 (1950).

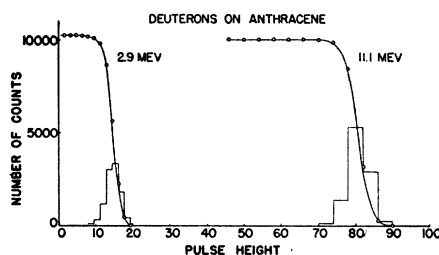


FIG. 2. Typical integral and differential pulse-height distribution curves for deuterons incident on anthracene.

<sup>4</sup> Harding, Flowers, and Eppstein, *Nature* **163**, 990 (1949).

<sup>5</sup> E. G. Michaelis, *Helv. Phys. Acta* **23**, Sup. 3, 155 (1950).

<sup>6</sup> J. B. Birks, *Proc. Phys. Soc.* **A63**, 1294 (1950) and private communication.

<sup>7</sup> Franzen, Peele, and Sherr, *Phys. Rev.* **79**, 742 (1950) and private communication.

<sup>8</sup> Frey, Grim, Preston, and Gray, *Phys. Rev.* **82**, 372 (1951).

<sup>9</sup> J. Broser and H. Kallmann, *Ann. Physik* **3**, 317 (1948).

<sup>10</sup> B. Collinge, E. J. Rolbins, *Nature* **166**, 1109 (1950).

<sup>11</sup> S. A. E. Johansson, *Ark. Fys.* **2**, 171 (1950).

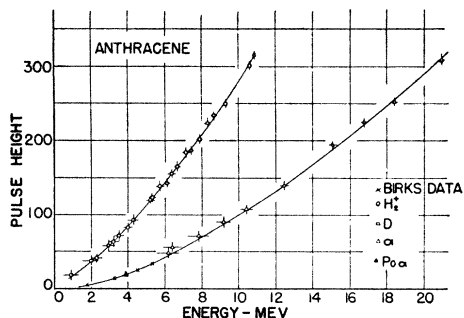


FIG. 3. Pulse heights resulting from the fluorescence radiation of anthracene when excited by heavy particles of different energies.

efficiency of crystals to electrons and heavy particles with the same specific energy loss. It was further desirable to obtain additional data to that already available on the response of various scintillation counters to electrons.<sup>12-14</sup> This would be necessary for the use of these detectors as beta- and gamma-ray spectrometers. With this intent, data were obtained on the response of the crystals to electrons in the low energy region (500 ev to 5000 ev) using a specially fabricated pulsed electron gun, while for a higher energy region (27 kev to 624 kev), the electrons were supplied by internal conversions and x-rays from radioactive sources.

## II. EXPERIMENTAL PROCEDURE FOR HEAVY CHARGED PARTICLES

The magnetically analyzed beam of the cyclotron was collimated by a set of brass slits 2 mm in diameter and brought out into air through a thin Nylon window (1 cm air equivalent) and allowed to strike the scintillation crystal under investigation. The crystal was mounted in optical contact with the photosensitive surface of an RCA 5819 photomultiplier tube and covered with a 0.16 mg/cm<sup>2</sup> reflecting aluminum foil as shown in Fig. 1.

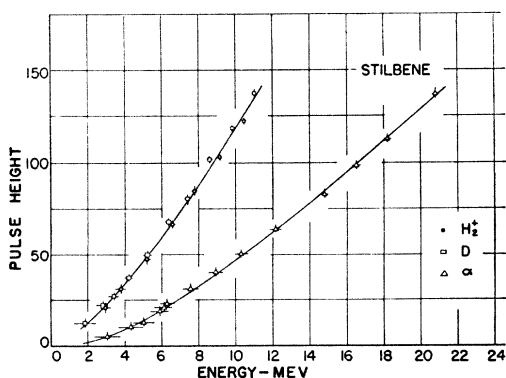


FIG. 4. Pulse heights resulting from the fluorescence radiation of stilbene when excited by heavy particles of different energies.

<sup>12</sup> J. I. Hopkins, Rev. Sci. Instr. 22, 29 (1951).

<sup>13</sup> West, Meyerhof, and Hofstadter, Phys. Rev. 81, 141 (1951).

<sup>14</sup> P. R. Bell, Science 112, 7 (1950).

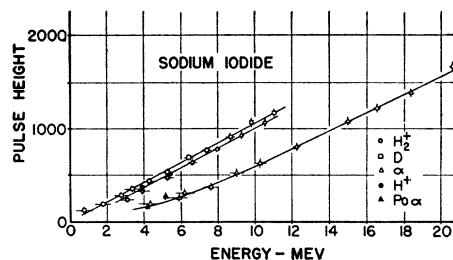


FIG. 5. Pulse heights resulting from the fluorescence radiation of thallium activated sodium iodide when excited by heavy particles of different energies.

An anthracene crystal 2.5×2.5×0.3 cm, a stilbene crystal 1×1×0.15 cm, and freshly cleaved sodium iodide crystals approximately 1.5×1×0.3 cm were used in obtaining the data to be described. The sodium iodide crystals were cleaved in a dry box, and the atmosphere inside the light-tight container was kept dry with calcium chloride. Frequent checks were made to guard against surface deterioration. Aluminum absorbers were interposed in the beam to vary the energy of the particles striking the crystal. The photomultiplier tube was magnetically shielded by four concentric cylinders of annealed mu-metal insulated from each other. An integrating time constant of 0.3 microsecond was used at the output of the photomultiplier tube for pulses from anthracene and stilbene, which have decay times of approximately 0.03 and 0.006 microsecond respectively,<sup>15</sup> while a time constant of 2.5 microseconds was used for pulses from sodium iodide, which has a decay time of approximately 0.25 microsecond.<sup>2</sup> These pulses were then applied to a cathode follower and a Los Alamos type model 100 linear pulse amplifier<sup>16</sup> and, after being shaped to flat-topped pulses

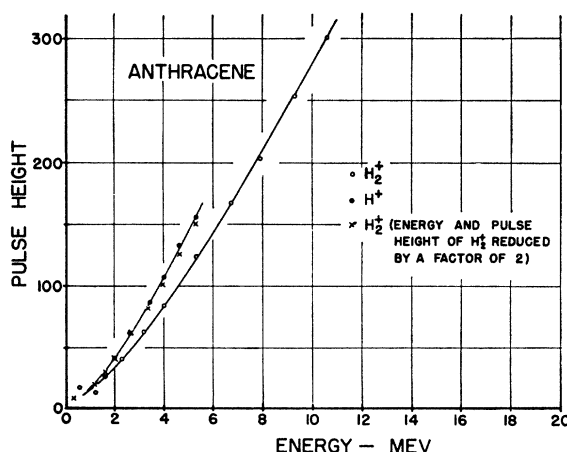


FIG. 6. Pulse heights resulting from the fluorescence radiation of anthracene when excited with molecular hydrogen ions and protons. A comparison is shown between the proton points and the H<sub>2</sub><sup>+</sup> points with the energies and pulse heights of the latter reduced by a factor of two.

<sup>15</sup> A. Lundby, Phys. Rev. 80, 477 (1950).

<sup>16</sup> W. C. Elmore and M. Sands, *Electronics* (McGraw-Hill Book Company, Inc., New York, 1949).

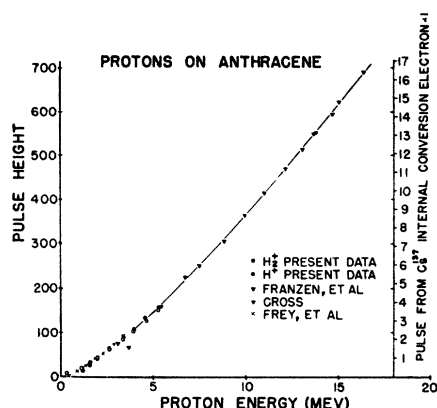


FIG. 7. Fluorescence radiation as a function of proton energy for an anthracene scintillation counter. The arbitrary ordinate scale on the left is the same as in Figs. 3-6, while the scale on the right is given in units of pulse height of the  $\text{Cs}^{137}$  conversion electrons.

7 microseconds wide, were analyzed with a twelve channel pulse amplitude analyzer using Atomic Instrument Company discriminators and scalars.

The cyclotron was operated with a very low beam to give approximately 100 counts per second from the scintillation counter. A total of 10,000 counts was taken at each energy.

The energy of the incident particles was determined under the same cyclotron conditions by measuring their ranges in air. This was done by passing the beam through a monitor consisting of a double-end-window proportional counter, whose stopping power was known, and then into a thin zinc sulfide screen placed on the end of a movable 5819 photomultiplier tube. This second detector had a threshold detection energy as measured with a polonium alpha-source of less than 30 kev. The energies obtained for protons, deuterons, and alpha-particles were  $5.76 \pm 0.06$  Mev,  $11.51 \pm 0.12$ , and  $22.87 \pm 0.23$  Mev respectively.

After emerging from the Nylon window, the particles from the cyclotron passed through 1.5 cm of air, through the aluminum absorber, and then through 3.5 cm of air before striking the crystal (see Fig. 1). The energy of the particle striking the crystal was then calculated using Bethe's range-energy curves<sup>17</sup> for the

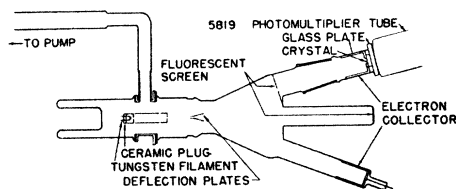


FIG. 8. Vacuum tube showing electron gun, crystal position, and electron collectors as used for the electron beam experiments.

<sup>17</sup> H. A. Bethe and L. M. Brown, private communication; H. A. Bethe, *Revs. Modern Phys.* **22**, 213 (1950).

two air paths and values from Smith<sup>18</sup> and University of California<sup>19</sup> for the aluminum absorbers.

The degradation of the energy by the aluminum foils to obtain the lower energies resulted in somewhat less energy resolution in the low energy region due to straggling. However, calculations showed that the effects of single and multiple scattering on the energy resolution were insignificant.

### III. RESULTS FOR HEAVY CHARGED PARTICLES

Typical pulse-height distributions are shown in Fig. 2. The differential histograms obtained essentially fit gaussian distributions with full widths at half-maxima of six to eight percent for the higher energy particles

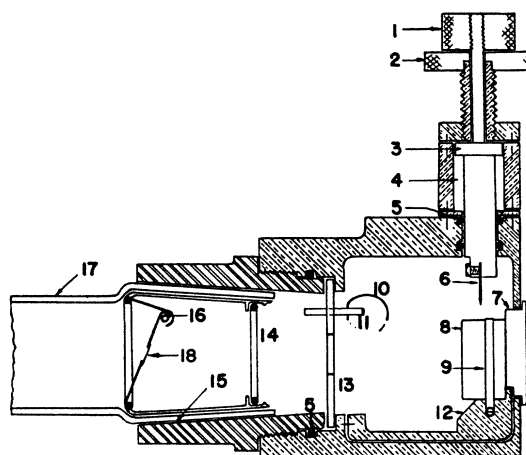


FIG. 9. Chamber used to cleave the sodium iodide crystals in the vacuum. 1,2-knurled knobs, 3-key, 4-keyway, 5-"O" ring, 6-razor edge, 7-Lucite window, 8-NaI crystal, 9-aluminum strap, 10-aluminum shield, 11-polonium alpha-source, 12-removable cradle, 13-aluminum plate, 14-piano wire spring, 15-34/45 standard taper for a greased joint. 16-mu metal tip. 17-Pyrex arm of vacuum tube. 18- $\text{Cs}^{137}$  source. Optical contact is made between the crystal and the Lucite window by means of a thin layer of silicone high vacuum grease. Rotating knurled knob 2 forces the piston down and presses the razor edge 6 against the upper face of the crystal. A penetration of a few millimeters is sufficient to cleave and separate the crystal halves, the rear half of which falls to the lower step of the cradle. The  $\text{Cs}^{137}$  source is mounted on a mu metal tipped aluminum arm and is positioned by an exterior magnet.

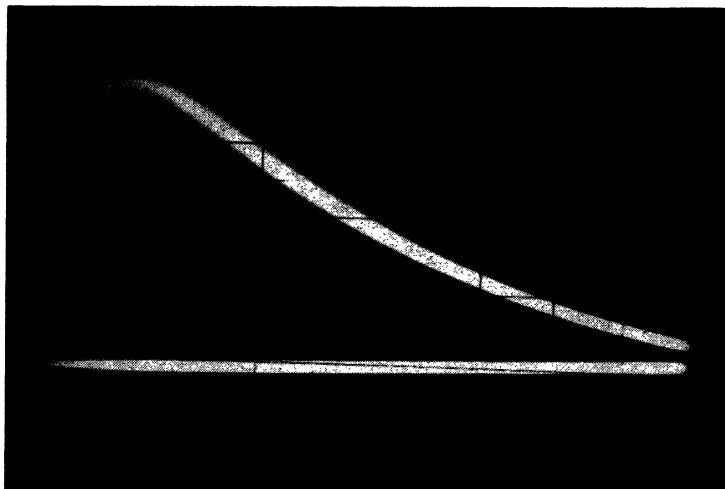
depending on the type of crystal. Although these widths were inherent in the gaussian plots, the peak pulse heights at various energies could be determined to within  $\pm 2.0$  percent with increasing uncertainties at the lower energies. This error was ascertained from the deviations in three complete energy response curves for deuterons on stilbene.

The variation of the mean pulse height with energy for the different particles and crystals used is shown in Figs. 3-5. The pulse-height scale is common to the three figures. The vertical lines indicate estimates of

<sup>18</sup> J. H. Smith, *Phys. Rev.* **71**, 32 (1947).

<sup>19</sup> University of California Radiation Laboratory Report 121, Revised November 1948.

FIG. 10. Pulses resulting from the fluorescence radiation of anthracene when excited by 5000 ev electrons with approximately 300 electrons in each pulse.



the pulse-height errors. The major error in the energy is introduced by the initial uncertainty of one percent in the energy. As the energy of the particles is reduced by the introduction of absorbers, the initial one percent error produces progressively larger uncertainties for the lower energy particles. The horizontal lines attached to the points show the total variation in energy at a given energy corresponding to a variation of one percent in the initial energy. These are not to be interpreted as errors in the determination of the energies, but rather as the amount by which the entire curve would be translated in energy for a one percent change in the initial energy. Errors introduced in the determination of an energy corresponding to a given aluminum absorber are certainly less than 0.1 Mev.

Although two curves were drawn for the deuteron and molecular hydrogen ion points for all three crystals in a preliminary report of these data,<sup>20</sup> further investigation has shown that the points for the organic crystals should all essentially lie on one curve for each type crystal, as in Figs. 3 and 4. The apparent small difference in the fluorescence radiation from sodium iodide when excited by deuterons and  $H_2^+$  ions is not understood.

The figures show a distinct difference in the behavior of the organic crystals and sodium iodide. The organic

TABLE I. Sources of internal conversion electrons used for single electron pulses.

Source	Energy of K-electron kev	Energy of L-electron kev	K : L ratio
Nb <sup>91</sup> a	86.9	103.0	2.5:1
In <sup>114</sup> b	164.1	187.7	1:1
Hg <sup>203</sup> b	196	264	3:1
Cs <sup>130</sup> c	623.9	655	5:1 <sup>b</sup>

<sup>a</sup> J. Ovadia, University of Illinois Ph.D. thesis, 1951.

<sup>b</sup> Nuclear Data, National Bureau of Standards Circular 499 (1950).

<sup>c</sup> L. M. Langer and R. D. Moffat, Phys. Rev. **78**, 74 (1950).

<sup>20</sup> Taylor, Remley, Jentschke, and Kruger, Phys. Rev. **83**, 169 (1951). Jentschke, Eby, Taylor, Remley, and Kruger, Phys. Rev. **83**, 170 (1951).

crystals have a nonlinear response of pulse height to energy in the entire region investigated. In sodium iodide protons and deuterons give linear response, while alpha-particles have a nonlinear relation over a low energy region with a linear response above an energy of about 10 Mev. The possibility of a deterioration of the crystal surface (due to the hygroscopic properties of sodium iodide) during a run which could contribute to this nonlinearity of response has been checked by experiments with crystals freshly cleaved in a vacuum. Results obtained in this way agreed with earlier ones, as indicated by the polonium alpha-point at 5.3 Mev in Fig. 5.

Figure 6 shows the response of anthracene to both protons and molecular hydrogen ions. It should be noted here that the  $H_2^+$  ions were actually detected as two protons entering the crystal simultaneously since the ions were dissociated when they passed through the Nylon foil. Hence if one divides both pulse height and energy of the  $H_2^+$  points by two, these reduced points should coincide with the proton curve as shown in the figure. Thus proton pulse height *vs* energy curves are readily obtained from the molecular hydrogen ion curves.

Using the response of the 624-kev internal conversion electrons<sup>21</sup> from Cs<sup>137</sup> as a normalization value,

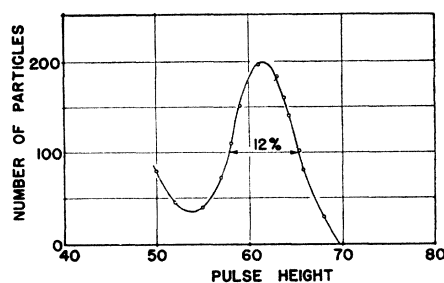


FIG. 11. Pulse-height distribution of the fluorescence radiation from Cs<sup>137</sup> conversion electrons incident on anthracene.

<sup>21</sup> L. M. Langer and R. D. Moffat, Phys. Rev. **78**, 74 (1950).

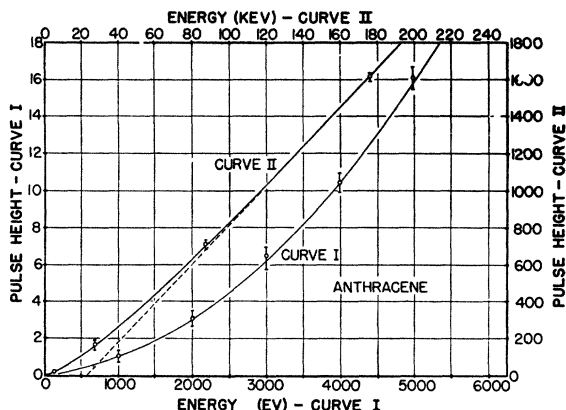


FIG. 12. Pulse heights from the fluorescence radiation of anthracene as a function of electron energy. The arbitrary ordinate scale must be multiplied by the factor 0.00673 to obtain the same scale used in Figs. 3-6 for the heavy particles.

the proton data for anthracene by Franzen, *et al.*,<sup>7</sup> the data of Frey, *et al.*,<sup>8</sup> some recent data by Cross,<sup>22</sup> and the above data have been plotted to the same scale giving the results shown in Fig. 7. The agreement of these sets of data is very satisfactory with the exception of the point of 3.7 Mev by Franzen, *et al.*,<sup>7</sup> the exact energy of which might be questioned as they indicate.

The ratio of the stilbene pulse height to the anthracene pulse height is closely the same for all three particles over the energy range investigated. This ratio is about 0.40 for  $H_2^+$  ions between 3 and 11 Mev, 0.40 for deuterons between 2 and 11 Mev, and 0.44 for alpha-particles between 5 and 21 Mev. Harding, *et al.*<sup>4</sup> have reported 0.5 for this ratio using 8.76-Mev alpha-particles.

Michaelis<sup>5</sup> gives pulse heights obtained for alpha-particles with energies of 3.1, 3.75, and 4.45 Mev incident on sodium iodide. Unfortunately, the amount of overlap with the results of this investigation is small. However, the results of Michaelis fit reasonably well on

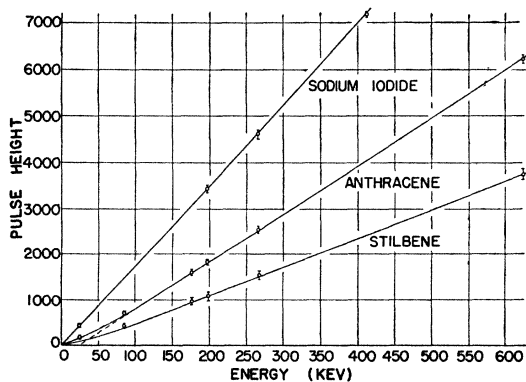


FIG. 13. Pulse heights from fluorescence radiation of anthracene, stilbene, and sodium iodide as a function of electron energy. The ordinate scale is the same as in Fig. 12.

<sup>22</sup> W. G. Cross, Chalk River Laboratory, private communication.

the low energy end of the observed sodium iodide curve when normalized at 4.45 Mev. The response of anthracene to alpha-particles in the energy range from 0.7 to 5.3 Mev as given by Birks<sup>6</sup> agrees well with the results obtained in this investigation as shown in Fig. 3. The results reported here for protons on sodium iodide show the same characteristic linearity as those of Franzen, *et al.*,<sup>7</sup> and appear to intersect very nearly at the origin.

#### IV. PROCEDURE FOR THE ELECTRON EXPERIMENTS

Since electrons with energies of 500 to 5000 eV produce ionization densities comparable to those produced by protons of two to fifteen Mev, the fluorescence of crystals when excited by electrons in this energy range was investigated. As it is practically impossible to measure the pulse heights of single electrons in this low energy region with any accuracy using organic crystals,<sup>23</sup> a pulsed monoenergetic electron beam was produced by a special cathode-ray gun for the examina-

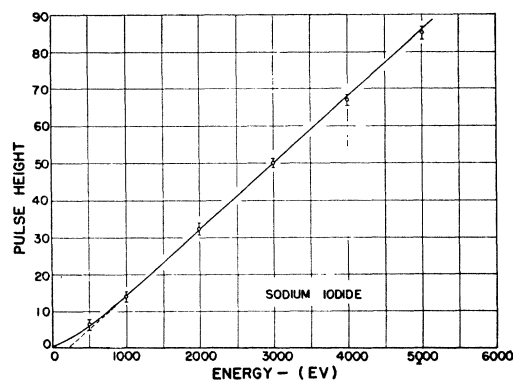


FIG. 14. Pulse heights from the fluorescence radiation of sodium iodide as a function of electron energy. The ordinate scale is the same as in Figs. 12 and 13.

tion of scintillations produced by electrons of these energies. This apparatus, shown in Fig. 8, produced an electron beam which impinged on the crystals after being deviated by the deflecting plates of the electron gun. This was done to effectively shield the crystal and photomultiplier tube from light produced by the filament of the gun. The magnetically shielded RCA 5819 tube was placed with the photocathode in contact with the glass plate on which the crystal was mounted at the end of the vacuum tube. Crystals  $1\text{ cm} \times 1\text{ cm} \times 0.25\text{ cm}$  were mounted in the apparatus, as indicated in Fig. 8. A vibrating reed electrometer,<sup>24</sup> with which the average beam current was measured, was connected to the electron collector.

The electron beam was pulsed at a rate of ten thousand per second with pulse durations of 0.5 and 2.0 microseconds. Identical results were obtained with

<sup>23</sup> W. J. Ramler and M. S. Freedman, *Rev. Sci. Instr.* **21**, 784 (1950).

<sup>24</sup> Palevsky, Swank, and Grenchik, *Rev. Sci. Instr.* **18**, 298 (1947).

both types of pulses indicating that no long-lived fluorescence of appreciable intensity could have been present. The average electron beam current was maintained at less than  $5 \times 10^{-13}$  ampere, so that no more than 300 electrons were incident on the crystal during any one pulse. Since the electron beam was defocused over an area of four mm<sup>2</sup> on the crystal, the particle density was small enough that the mechanism for producing fluorescence was assumed to be the same as that for single electrons, and hence a pulse height vs energy relation for single electrons could be obtained.

The secondary emission coefficients of the crystals used are greater than one for the electron energies investigated, so that no significant charge collection should occur on these insulating crystals. As a check the

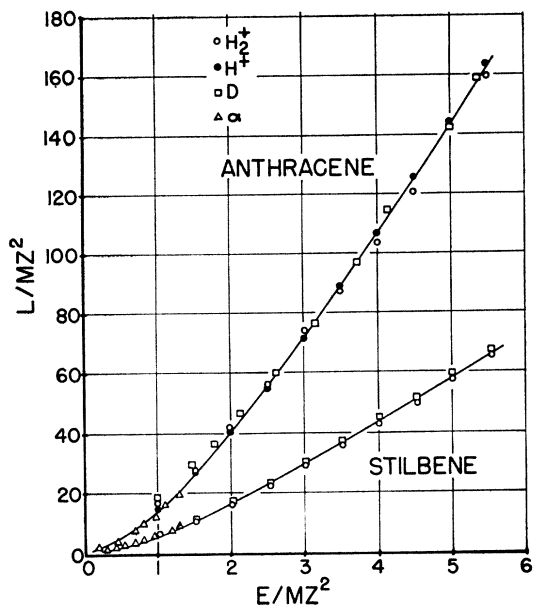


FIG. 15. Fluorescence radiation of anthracene and stilbene when excited by heavy particles of different energies with ordinates and abscissas reduced by the factor  $1/MZ^2$ ;  $M$  and  $Z$  are mass and charge of the incident particle respectively.

current could also be measured with a similar charge collector made completely of tungsten, and shown in the lower arm of the vacuum tube in Fig. 8. No difference in the current measurements could be detected if a sufficiently large potential (25 volts) was applied to this second collector to prevent the escape of an appreciable number of secondary electrons.

The proper performance of the apparatus could be checked by a variation of the beam intensity. Varying the electron current by a factor of ten produced a proportionate change in the output pulses of the photomultiplier.

The effect of the back diffusion<sup>25</sup> of the electrons striking the crystal restricts the accuracy of the results achieved with the electron gun. Extrapolating the

<sup>25</sup> W. Bothe, Handbuch der Physik 22, Part 2, 23 (1933).

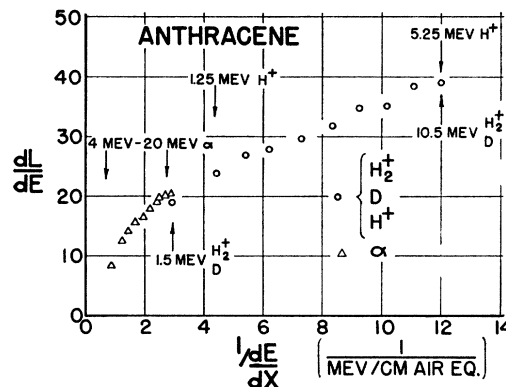


FIG. 16. The differential efficiency of fluorescence,  $dL/dE$ , for anthracene as a function of the reciprocal of the specific energy loss,  $dE/dx$ , of the incident heavy particles of different energies.  $dL/dE$  is found from the curves in Fig. 3 and is expressed in arbitrary units per Mev, where the arbitrary units are the same as the ordinate scale in Fig. 3.

results of Schonland<sup>26</sup> to the low energy region and to elements of low atomic number, the ratio of the number of electrons which escape by back diffusion to the number incident should be less than 10 percent for the organic crystals, but the ratio could be as large as 30 percent for sodium iodide.

In order to normalize the data taken with the electron gun to other data, a small source of  $Cs^{137}$  on a 3.2 mg/cm<sup>2</sup> aluminum foil was mounted in such a manner that the conversion electrons were incident on the crystals at the same spot as the electron beam. During the electron beam runs, the radioactive source was moved out of the beam path with a magnet.

Since sodium iodide is quite hygroscopic, a special chamber was constructed for the experiments with these crystals. This chamber, shown in Fig. 9, permitted

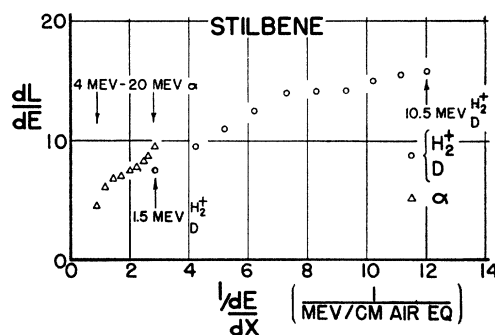


FIG. 17. The differential efficiency of fluorescence,  $dL/dE$ , for stilbene as a function of the reciprocal of the specific energy loss,  $dE/dx$ , of the incident heavy particles of different energies.  $dL/dE$  is found from the curves in Fig. 4. The ordinate scale is the same as Fig. 16.

<sup>26</sup> B. F. J. Schonland, Proc. Roy. Soc. (London) A104, 235 (1923); 108, 187 (1925). He has shown that for a parallel beam of electrons normally incident on the surface, the ratio of back scattered to incident electrons is practically energy independent within the investigated region of 10 to 70 kev. Hence our extrapolation to lower energies seems to be justified.

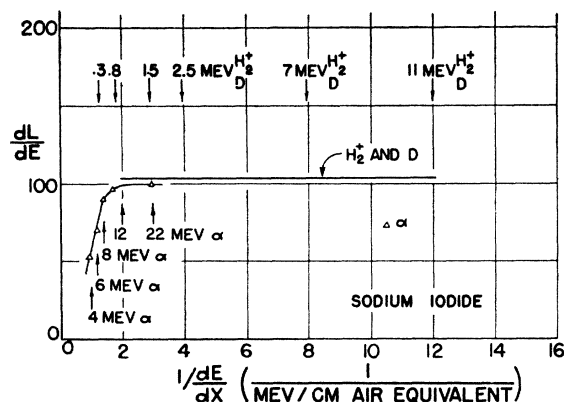


FIG. 18. The differential efficiency of fluorescence,  $dL/dE$ , for sodium iodide as a function of the reciprocal of the specific energy loss,  $dE/dx$ , of the incident heavy particles of different energies.  $dL/dE$  is found from the curves in Fig. 5. The ordinate scale is the same as Figs. 16 and 17.

the installation of a crystal in the vacuum associated with the gun. The crystal could then be cleaved in the vacuum just prior to taking data, and no deterioration of the crystal surface could occur.

Positive pulses were taken from the photomultiplier tube, amplified by a linear amplifier, and photographed on a Tektronix high speed oscilloscope.

In order to extend the data for these low energy electrons to higher energies, internal conversion electrons from radioactive sources were used. These artificial sources are summarized in Table I. Since there were "Auger electrons" present in the radiation striking the crystal, the distance from the source to the crystal was kept at greater than one cm to minimize the simultaneous detection of a conversion electron with an Auger electron. The  $K$  and  $L$  conversion lines could not be resolved for the  $Nb^{91}$  and  $In^{114}$  sources, so the points from these are given for a mean energy of the two lines considering their known intensities. The  $Hg^{203}$  lines were easily resolved giving two points, and the  $L$  line of  $Cs^{137}$  was too weak to give a point with sufficient accuracy. An intermediate energy was obtained using

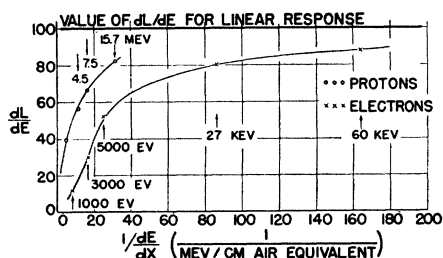


FIG. 19. The differential efficiency of fluorescence,  $dL/dE$ , for anthracene as a function of the reciprocal of  $dE/dx$  of the incident protons and electrons of different energies. The arbitrary ordinate scale has been chosen so that 100 is the value of  $dL/dE$  resulting from electrons of sufficiently high energy (above 100 kev) to give linear response. The efficiency for conversion of incident particle energy into light energy attains its maximum value here and amounts to about 2 or 3 percent.

the 27-kev x-rays from  $L$  to  $K$  electron transitions in  $Te^{127}$ . Using the same type optical arrangement as for the heavy particles, the pulses from these single electrons were amplified and analyzed with the twelve channel pulse amplitude analyzer. In these measurements with the higher energy single electrons back diffusion of the electrons, though present, did not affect the data since the histograms of the pulse-height distributions of the conversion electrons appeared to be symmetrical about the maximum.

## V. RESULTS OF THE ELECTRON EXPERIMENTS

A typical photograph of the pulse height resulting from 5000-ev electrons incident on anthracene is shown in Fig. 10. The pulse-height distribution from the cesium electrons in anthracene, as taken with the counting technique, is given in Fig. 11.

The pulse height *vs* energy curves for the electrons in the various crystals are given in Figs. 12–14. The ordinates of each point represent the mean pulse height resulting from six measurements at each energy. The vertical lines indicate the largest deviations of these individual measurements.

Anthracene and stilbene show a nonlinear response below 100 kev in agreement with Hopkins' investigation of anthracene.<sup>12</sup> The linear portions of these curves above 100 kev extrapolate to an intercept of about 22 kev on the energy axis.

Sodium iodide shows a linear response throughout the energy region from 1 kev to 624 kev in agreement with the data from the Stanford group,<sup>13</sup> who find linearity above 2 kev. Below 1 kev there are indications of a deviation from linearity as shown in Fig. 14, which might be explained by back diffusion effects.

## VI. DISCUSSION OF RESULTS

It is well known that the fluorescence efficiency of a crystal is dependent on the current density of the incident particles. It has also been suggested, probably first by Kallmann,<sup>27</sup> that the fluorescence efficiency of a crystal for single particles is dependent on the density of ionization along the path of the particle in the crystal. We are indebted to Professor F. Seitz for the following formal approach to a possible explanation of the difference in response of a single organic crystal to different heavy particles. If one assumes that the differential fluorescence efficiency,  $dL/dE$ , for producing fluorescent radiation depends on the specific energy loss of the particles,  $dE/dx$ , then one can write:

$$dL/dE = e = f(dE/dx) \quad (1)$$

or

$$dL/dx = e(dE/dx) = (dE/dx)f(dE/dx). \quad (2)$$

The expression for the specific energy loss as given by

<sup>27</sup> H. Kallmann, *Z. Naturforsch.* **3a**, 6 (1948); H. Kallmann, Conference on Scintillation Counters, Oak Ridge, Tennessee, June, 1949.

Livingston and Bethe<sup>28</sup> can be written as:

$$-(dE/dx) = (2\pi Z^2 z e^4 N M / m E) \ln(4mE/MI) \quad (3)$$

where  $Ze$ ,  $M$ , and  $E$  are the charge, mass, and energy of the incident particle and  $Nz$ ,  $m$ , and  $I$  are the number of electrons per cm<sup>3</sup> of the stopping material, the mass of the electron, and the average excitation potential of the atom of stopping substance. For a given crystal  $N$ ,  $z$ ,  $m$ , and  $e$  are constant and the logarithm term is practically constant—varying between 3.3 and 5.1—for the particles and energies used. Therefore to a good approximation, Eq. (3) can be written as:

$$dE/dx = (MZ^2/E)B \quad (4)$$

where  $B$  is essentially constant. Then we can express:

$$dL/dE = g(E/MZ^2) \quad (5)$$

whence the total light output for an incident particle of energy  $E$  is:

$$L = \int_0^E g(E'/MZ^2) dE' \\ = MZ^2 \int_0^{E/MZ^2} g(E'/MZ^2) d(E'/MZ^2) \quad (6)$$

or

$$L/MZ^2 = h(E/MZ^2), \quad (7)$$

where  $h$  is a function of  $E/MZ^2$ . Therefore for a given crystal, the curves for different incident particles should reduce to a common curve when  $L/MZ^2$  is plotted vs  $E/MZ^2$ . Such curves are shown in Fig. 15 for the anthracene and stilbene data.

It appears that the curves obtained for the different heavy particles do reduce to a common curve for each crystal. However, the alpha-particle curve is compressed to the extent that it lies at the extreme low energy end of the proton and deuteron curves, so that much higher energy alpha-particles would be required for a more thorough check. The data are somewhat insensitive to this approach, but they do support the assumption that, for heavy particles, the fluorescence efficiency is dependent on the specific energy loss in the crystal, which is proportional to the ionization density.

A more sensitive dependence to the rate of energy loss,  $dE/dx$ , can be presented by plotting the fluorescence efficiency,  $dL/dE$ , obtained by taking the slopes of the curves in Figs. 3-5, as a function of  $dE/dx$ . The resulting curves for the heavy particles are shown in Figs. 16-18, where the fluorescence efficiency is plotted vs the reciprocal of  $dE/dx$ . The specific energy loss has been calculated for these curves from the range-energy data of Bethe.<sup>17</sup> If the response of the crystals were proportional to the energy of the particles, the efficiency would be a constant, independent of the energy. For the organic crystals  $dL/dE$  increases

<sup>28</sup> M. S. Livingston and H. A. Bethe, *Revs. Modern Phys.* 9, 263 (1937).

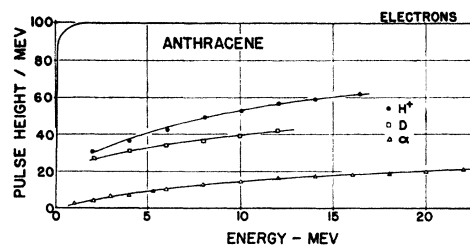


FIG. 20. Ratios of the total fluorescence radiation from anthracene for different particles to the energies of the particles plotted as functions of the energies. The arbitrary ordinate scale has been chosen so that the maximum value for electrons is 100 units per Mev.

continuously with increasing values of  $1/dE/dx$ , with the proton and deuteron values lying closely together, while the points from the alpha-particle data appear to be consistent with a continuation of the proton and deuteron points. However it is possible that the alpha points lie slightly above the continuation. If this is true, it could be a result of the different energy spectrum of the secondary electrons from alpha-particles as compared to the secondary electrons from protons and deuterons of the same specific energy loss. Since the secondaries from the alpha-particles have greater energies, the ionization density along the paths of the particles will be less, and this will result in a larger value of  $dL/dE$  for a given value of  $dE/dx$ .

For sodium iodide the fluorescence efficiency for protons and deuterons is constant, as would be indicated by the linear response to those particles. The deviations from linearity for the alpha-particles—the variable portion of the curve in Fig. 18—occur at values of  $dE/dx$  corresponding to  $H_2^+$  and deuteron energies of about 0.4 Mev, which are too small for any region of nonlinear response to be detected.

The fluorescence efficiency vs the reciprocal of the specific energy loss for electrons and protons in anthracene is compared in Fig. 19. The values of  $dL/dE$  for the higher proton energies were obtained from the data of Franzen, *et al.*<sup>7</sup> The values of  $dE/dx$  for electrons

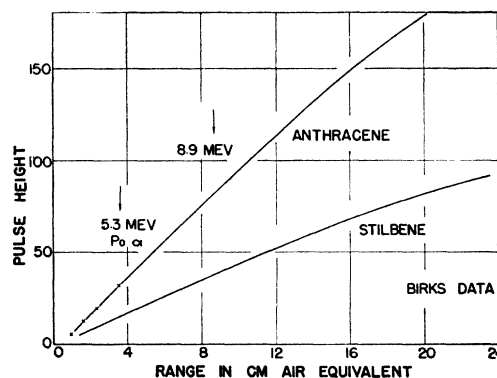


FIG. 21. Pulse heights resulting from the fluorescence radiation of anthracene and stilbene when excited by alpha-particles plotted as a function of the range of the particles in air. The ordinate scale is the same as in Figs. 3-6.



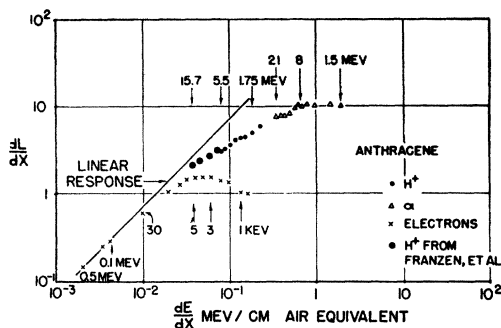


FIG. 22. Variation of the specific fluorescence,  $dL/dx$  (expressed in arbitrary units per cm of air equivalent), of anthracene with the specific energy loss,  $dE/dx$ , of the incident alpha-particles, protons, and electrons of different energies. The straight line indicates linear response of the crystal.

were calculated for anthracene from Bethe's theoretical formula<sup>29</sup> using a calculated value of 1200 as a mean value for the stopping power of anthracene relative to air. Since the energy loss per ion is about the same for both protons and electrons, one might expect that the two curves would roughly coincide if the fluorescence efficiency is dependent only on the ionization density. As is shown in Fig. 19, the curves certainly are not coincident, but the values of  $dL/dE$  for the higher proton energies appear to be approaching about the same constant value attained by  $dL/dE$  for electrons of energies sufficiently high enough to give linear response; i.e., above 100 keV. It should be noted, however, that the constant value of  $dL/dE$  for protons is perhaps reached at a considerably different value of  $dE/dx$  than for the electrons. If one considers the loss of pulse height due to the back diffusion of the electrons in the electron gun experiment, the actual difference between the two curves in the low energy region should be smaller than the measured difference. However if the estimated numbers of back scattered electrons (as given in Sec. IV) are correct, the difference in the curves cannot be explained by this effect alone, particularly since the electrons lost by back diffusion lose some energy in the crystal and thus make a small contribution to the pulse height. Since no accurate data applicable to our experimental conditions are available the back diffusion effect needs further investigation.

In sodium iodide, the electrons give the same values of  $dL/dE$  as the protons and deuterons as might be indicated by the linear response to all three particles. Here the back diffusion effect for the electron gun experiments should not be negligible. Nevertheless, the measurements of the relative pulse heights for the different electron energies should be accurate, since the amount of back diffusion in heavy elements is certainly independent of the electron energy. The fact that the absolute values of the pulse heights obtained with the electron gun measurements do compare so favorably

with the measurements taken with the single electrons is not understood.

A useful presentation of the data for anthracene is shown in Fig. 20, where the ratio of total pulse height at a given energy divided by that energy is plotted as a function of the energy for the particles investigated. The arbitrary ordinate scale has been chosen so that the maximum value for electrons is 100, this being attained in the region of linear response for the electrons. Thus the heavy particle curves give the response in percentage of the maximum light output per Mev.

For specific energy losses of heavy particles greater than 0.63 Mev/cm air equivalent, as in our case for alpha-particles below nine Mev, the specific fluorescence (the light output per cm air equivalent) attains a constant value in the organic crystals and is then independent of the specific energy loss. This is indicated in Fig. 21, in which plots of pulse height *vs* range in air of alpha-particles incident on anthracene and stilbene are shown. The curves are essentially linear up to a range of nine cm, and thus  $dL/dx$  is constant up to this range, where  $dE/dx$  is greater than the above mentioned value.

This effect in anthracene was first noted by Birks<sup>6</sup> and has been studied by Cross<sup>30</sup> using alpha-particles from ThC', ThC, Cm, Am, Pu, and from nuclear reactions. Reynolds, Harrison, and Hill<sup>31</sup> have found a similar saturation effect using liquid scintillation substances. It is interesting to note that in all the results mentioned here the linear portion of the line does not extrapolate through the origin.

Similarly, if the same types of curves are plotted for electrons on anthracene and stilbene the same results seem to be indicated, namely, a region where the curve is linear and hence  $dL/dx$  is constant, but the saturation of  $dL/dx$  is attained at a much smaller value of  $dE/dx$  than in the case of heavy particles. However due to the uncertainty of the correction for the back diffusion no definite conclusion can be made.

A survey of the response of the anthracene crystal to the different ionizing radiations is presented in Fig. 22, where the variation of the specific fluorescence is given as a function of the specific energy loss in a log-log plot. This curve seems to show the following trends. Both the heavy particles and electrons apparently give regions of linear response (the specific fluorescence increasing linearly with energy) but these probably starting at quite different values of  $dE/dx$ . An extrapolation is required to reach this linear region for the heavy particles. At larger values of  $dE/dx$ , but different for the heavy particles and electrons, the response becomes nonlinear for both types of particles, and at even larger values of the specific energy loss, the specific fluorescence for

<sup>30</sup> W. G. Cross, Chalk River Laboratory, private communication.

<sup>31</sup> Reynolds, Harrison, and Hill, Phys. Rev. **82**, 317 (1951), and private communication.

<sup>29</sup> H. A. Bethe, Handbuch der Physik **24**, Part 1, 521 (1933).

heavy particles saturates. The same effect is indicated for electrons, but due to the uncertainty of the magnitude of the back diffusion effect in this case no quantitative statement can be made. However, we do not believe that the different results for electrons and heavy particles could be a result of surface effects and back diffusion alone, since the experiments with the 27-kev x-rays, which produced electrons inside the crystal, gave data in agreement with the above results. §

§ *Note added in proof*:—J. B. Birks, *Phys. Rev.* **84**, 364 (1951), has recently used the exciton theory to explain the general features of these results.

## VII. ACKNOWLEDGMENTS

The authors are grateful to Professor F. Seitz for stimulating discussions concerning the interpretation of the data. They wish to thank A. L. Atkins, R. O. Kerman, and W. E. Kreger who determined the cyclotron beam energy. The valuable assistance of Mr. Chester Lob and the use of facilities of the Tube Laboratory of the Electrical Engineering Department in performing the electron experiments are deeply appreciated. The authors also would like to thank W. G. Cross of Chalk River Laboratory who kindly sent them his data prior to publication.

## Investigation of the Gliding Process in Ionic Crystals by Prismatic Punching

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(Received June 28, 1951)

The prismatic punching method applies a unidirectional stress on a very small surface (approximately 0.001 square centimeter) of a crystal lying on a support which is softer than the crystal itself. The method is very convenient for low and high temperature and for small samples. The glide system of thallium bromiodide crystals determined by the prismatic punching method contains dodecahedron planes (110) as glide planes and cubic planes [001] as glide directions. The width of the glide bands ranges from 1 to 5 microns. The stress-strain curve shows a linear relation between the shear angle  $\delta$  and the applied stress  $\sigma$  ( $\delta = a'\sigma + b$ ). The creep takes place according to the exponential law ( $\delta = at^m$ ). The glide bands have to be displaced a distance of about 100 ions at room temperature and about 15 ions at  $-190^\circ\text{C}$  before rupture takes place (plastic limit). Prismatic punching on sodium chloride crystals produces gliding along dodecahedron planes (110) in  $[1\bar{1}0]$  direction. Apparent cleavage of sodium chloride crystals along dodecahedron planes and normal cleavage along cubic planes are explained as ruptures between neighboring glide planes.

### I. INTRODUCTION

GLIDING is the basic process in the plastic deformation of crystals. Although numerous investigations have been made in this field, many problems are still unsolved. There exists a great need for new experiments which may contribute to their solution. Great progress has been achieved in the study of the gliding process, mainly by investigations of single metal crystals.<sup>1</sup> Nonconducting crystals as compared with metals have some advantages. Since most of them are transparent, they permit study of the gliding process in their interior by observation between crossed polarizers.<sup>2</sup> On the other hand, nonconducting crystals have the disadvantage of a very narrow plastic range; therefore, the method for studying the gliding process must be very sensitive. However, there exist a few ionic crystals which possess plastic properties resembling metals. Those crystals are the silver and thallium halides

which can be easily grown in large sizes. As pure silver and thallium halides are very soft, they require special care when specimens are being prepared. Mixed crystals of thallium halides are almost ideal for gliding studies as their hardness is considerably greater than that of their pure components. Thallium bromiodide crystals of the composition 41.7 percent TlBr + 58.3 percent TlI by weight have been used in this investigation.

The methods generally used for investigation of the gliding process are based on the study of shear stress-strain curves. The simplest conditions exist when pure shear stress is applied. Under tension or volumetric compression, however, no pure shear stress is present. The same is true of torsion method. The Bausch method,<sup>3</sup> which claims to give a unidirectional stress, gives no uniform shear stress, as has been shown by Read.<sup>4</sup> In addition, all these methods require large crystal specimens which are seldom obtainable; other disadvantages which should also be mentioned are the variable influence of the specimen surface on the stress-strain and the multiple gliding along different glide planes.

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<sup>1</sup> For general summary and references see: *A Symposium on the Plastic Deformation of Crystalline Solids*, Mellon Institute, Pittsburgh 19, 20 May 1950.

<sup>2</sup> E. Schmidt and W. Boas, *Kristallplastizität* (Verlag-Springer, Berlin, 1935).

<sup>3</sup> K. Bausch, *Z. Physik.* **93**, 479 (1935).

<sup>4</sup> W. T. Read, see reference 1, p. 111.

FIG. 10. Pulses resulting from the fluorescence radiation of anthracene when excited by 5000 ev electrons with approximately 300 electrons in each pulse.

