The Detection of Gamma-Rays with Thallium-Activated Sodium Iodide Crystals*

ROBERT HOFSTADTER
Palmer Physical Laboratory, Princeton University, Princeton, New Jersey
(Received September 27, 1948)

Thallium-activated sodium iodide crystals may be used as efficient detectors of gamma-rays and other ionizing radiations. The crystals are used in combination with a photo-multiplier after the manner of Kallmann. Curves are given which show the duration of the light emission process, the distribution of light pulse sizes, the energy discrimination possibilities, and the dependence of number of counting events on photo-multiplier voltage and amplifier gain. Comparison curves are shown for NaI(Tl) and anthracene. A description is given of the method of preparation of polycrystalline samples and single crystal specimens of the sodium iodide-thallium phosphor. A few remarks are made concerning the combination of alkali halide phosphor crystals with photographic plates.

I. INTRODUCTION

THE detection of beta-particles and gamma-rays by the combination of a luminescent crystal (phosphor) and a photo-multiplier vacuum tube has been successfully demonstrated by many investigators.[1–6] The earliest work in which large crystalline samples were used was performed by H. Kallmann.[1] Kallmann showed that large clear pieces of napththalene produced measurable scintillations when exposed to gamma- and beta-radiation from radium. Other luminescent materials have since been reported in the literature. Among these are anthracene,[5] calcium tungstate,[6] phenanthrene,** and others.

Since present-day photo-multiplier tubes have a serious "noise" background at room temperature,[7] it is important that the phosphor provide as large a light flash as possible. Other important properties of a useful phosphor are: the spectrum of the light flash, the duration and rise of the light pulse, the material density, and, finally, the chemical composition of the phosphor. The magnitude (and spectrum) and duration of the light flash are of special significance since these properties determine the signal to noise ratio.

Amplified noise pulses are, unfortunately, not small compared with light flashes in naphthalene, calcium tungstate, and anthracene. Consequently, there are important advantages in cooling the photo-multiplier tube to low temperatures at which the frequency of occurrence of noise pulses becomes insignificant compared with the signal. On the other hand, the experimental difficulties in cooling the tube may prove to be a disadvantage in some experiments.

Of those phosphors mentioned above, anthracene produces relatively large light flashes[5] in short times[8] (~0.05-microsecond duration) and matches, in its emission, the region of high spectral sensitivity of the 931 or 1P21 photo-multiplier tubes. With respect to the remaining above-mentioned characteristics of useful phosphors, anthracene has a small density (1.25 g/cm^3) and is composed of elements of low atomic number (1, 6). Anthracene light flashes for, say, 1.0-Mev gamma-rays are not many times the size of photo-multiplier noise pulses, and, for a reason not yet understood, it appears that heavy particle pulses produce smaller scintillations than beta-particles of equivalent energy.[5] It is, therefore desirable to search further for a phosphor, more dense than anthracene, having constituents of higher atomic number, which provides larger light flashes than anthracene. It would also be desirable if such a phosphor would give large light flashes for heavy particles, such as alpha-particles. At the present time no phosphor has been reported which can be ob-

* The major part of these results was presented at a recent conference (July, 1948), on high speed counters, at the University of Rochester.
[1] H. Kallmann, Natur and Technik (July, 1947).
[2] J. W. Coltman and F. H. Marshall, Phys. Rev. **72**, 528 (1947).
[3] M. Deutsch, Nucleonics **2**, 58 (1948).
[4] G. B. Collins and R. C. Hoyt, Phys. Rev. **73**, 1259 (1948).
[5] P. R. Bell, Phys. Rev. **73**, 1405 (1948).
[6] R. J. Moon, Phys. Rev. **73**, 1210 (1948).
** M. Deutsch: Rochester Conference.
[7] R. W. Engstrom, J. Opt. Soc. Am. **37**, 420 (1947).
[8] L. F. Wouters, Phys. Rev. **74**, 489 (1948); G. B. Collins, Phys. Rev. **74**, 1543 (1948).

tained in large crystals, and which possesses the desirable characteristics discussed above.

It is the purpose of this article to present some of the properties of a material which is an efficient phosphor for alpha-, beta-, and gamma-radiations.[9] It has been found that sodium iodide, with a small thallium iodide impurity, possesses many desirable properties of a radiation detector. In the case of NaI(Tl) the light flashes are sufficiently large so that in most instances (e.g., 0.5-Mev gamma-rays) cooling of the photomultiplier is unnecessary. The density of NaI is 3.67 grams/cc, and a medium atomic number ($I=53$) is available in the phosphor. Large clear crystals can be prepared by relatively simple means. These features make the use of NaI(Tl) quite convenient for the counting of gamma- and beta-radiation. It is also true that heavy particle pulses are large. It cannot be said that NaI(Tl) has all the desirable properties of a phosphor which might be used for the general counting and measuring of ionizing radiation, for, as it will be shown, NaI(Tl) is not as fast as the organic phosphor materials. A practical disadvantage of NaI(Tl) is the fact that this material is highly hygroscopic, although this difficulty has been avoided by enclosure of the sample, as will be shown below. In summary, NaI(Tl), in its present form, would seem to be a useful material for alpha-, beta-, and gamma-radiations in those cases where resolving times are not required to be less than one or two microseconds.

In the following sections some experiences have been collected which concern the use of NaI(Tl) as a detector of gamma-rays. Data on the decay of luminescence of sodium iodide will be reported and some observations made on combinations of various phosphors and a photographic plate.

II. GENERAL

R. W. Pohl[10] and his co-workers have studied the alkali halide crystals with thallium impurities. In particular, this group has examined luminescence produced by ultraviolet light and the absorption of light in alkali halide phosphors. Much experimental and theoretical material on such phosphors now exists[11–15] for the significant reason that these phosphors are among the most simple now known. Nevertheless, the exact nature of the short-lived fluorescence is probably not completely understood at the present time. The thallous ion is probably responsible for the observed behavior.

In connection with the material of this paper, the experimental results of W. von Meyeren[16] and R. Hilsch[17] are of interest. These authors have studied the spectral distribution of the emitted fluorescent light, as well as absorption of light by the alkali halide-thallium phosphors. Absorption by the base materials has been studied by R. Hilsch and R. W. Pohl[18] and by E. G. Schneider and H. M. O'Bryan.[19] Unfortunately, NaI(Tl) has not been studied in emission, although its absorption is given by R. Hilsch.[11]

The experimental findings in some of the alkali halides (NaCl, KCl, RbCl, NaBr, KBr, KI) may be summarized in the following way. With a small (0.001 to 0.1 molar percent) thallium impurity these phosphors show luminescence lying in the near ultraviolet, and, in the case of KI, extending into the violet and blue region of the visible spectrum. With larger impurities (0.1 to 5 molar percent) the luminescence spectrum shifts towards the visible region.[12] With very large impurities the luminescent light

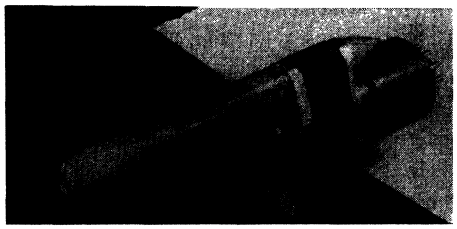

FIG. 1. Phosphor tube mounted on photo-multiplier.

[9] R. Hofstadter, Phys. Rev. **74**, 100 (1948).
[10] Bibliography in article by R. Hilsch (11).
[11] R. Hilsch, Proc. Phys. Soc. London **49**, extra part, 40 (1937).
[12] P. Pringsheim, Rev. Mod. Phys. **14**, 132 (1942).
[13] J. T. Randall and M. H. F. Wilkins, Proc. Roy. Soc. **184A**, 366 (1945).
[14] F. Seitz, J. Chem. Phys. **6**, 150 (1937).
[15] N. F. Mott and R. W. Gurney, *Electronic Processes in Ionic Crystals* (Oxford University Press, London, 1940), p. 224.
[16] W. von Meyeren, Zeits. f. Physik **61**, 329 (1930).
[17] R. Hilsch, Zeits. f. Physik **44**, 860 (1927).
[18] R. Hilsch and R. W. Pohl, Zeits. f. Physik **59**, 812 (1930).
[19] E. G. Schneider and H. M. O'Bryan, Phys. Rev. **51**, 293 (1937).

FIG. 2. Active region of tube of Fig. 1.

tends to be white. The time decay of the luminescent process varies also with the amount and kind of added impurity. There is a tendency to show a greater phosphorescent component as the amount of impurity increases. The above-mentioned emission of light is produced by stimulation by ultraviolet light lying in the absorption band or bands of the phosphors. The emitted light produced by ionizing radiation is probably, but not necessarily, the same as that produced by ultraviolet light.

The absorption of light by the alkali halide-thallium phosphors generally consists of the absorption by the base material in addition to the specific absorption bands introduced by the added impurity. The impurity absorption bands lie just beyond the emission bands on the side of shorter wave-lengths. It is fortunate that there is little overlap, and this fact makes the alkali halide phosphors quite transparent to their own fluorescent radiation. As an example, KI(Tl) absorbs in bands at 2870A and 2360A, whereas the emitted light (under ultraviolet excitation) extends from 3200A to 5000A. Such results have been stated for room temperature conditions. The emission of light has also been studied at temperatures above and below room temperature.[16] In the case of NaI(Tl), the absorption bands lie at 2930A and 2340A. The absorption of longer wave-lengths extends as far as 3200A.

Not much seems to be known of the period of decay of fluorescence in the thallium-activated alkali halide phosphors, although W. Bunger and W. Flechsig[20] have studied KCl with TlCl impurity and have found a decay at least as fast as 50 microseconds. W. Bunger[21] has observed high quantum efficiencies, of the order of 50 to 80 percent in KCl(Tl) phosphors, under excitation by ultraviolet light.

[20] W. Bunger and W. Flechsig, Zeits. f. Physik **67**, 42 (1931).

[21] W. Bunger, Zeits. f. Physik **66**, 311 (1930).

III. EXPERIMENTAL

A. Apparatus

In these experiments the photo-multiplier was mounted in a tube socket, on a small brass frame, within a light-tight galvanized iron box of $\frac{1}{32}$-inch wall thickness. The phosphor was enclosed in a quartz tube of $\frac{9}{16}$-in. outside diameter and 1-mm wall thickness. The quartz tube was placed, in a horizontal position, in contact with the photo-multiplier envelope. To prevent motion of the quartz tube, a length of adhesive tape was wrapped around the quartz tube and photo-multiplier envelope. A thin aluminum foil was placed between the sticky side of the tape and the quartz tube. In this way the phosphor tube could be removed easily and replaced easily. The aluminum foil also served as a reflector of light. Figure 1 shows the type of mounting used.

In the experiments to be described, the photo-multiplier was supplied with a stabilized negative voltage at the cathode. The anode load resistance was usually 0.1 megohm, although in some tests a 10.0-megohm resistance was employed. The combined capacitance of photo-multiplier output and amplifier input was 20 to 30 micromicrofarads. The time constant of the photo-multiplier output circuit was therefore of the order of two or three microseconds except in those cases where the 10.0-megohm load was used. The voltage divider supplying the dynodes was made up of resistances of value 33 kilo-ohms each. The anode circuit of the photo-multiplier was connected with a small lead directly to a preamplifier.

The amplifier used in most of these experiments has been the "Model 501" amplifier, designed by W. C. Elmore. It consists of a preamplifier, mentioned above, and a separate chassis containing the remaining amplifying stages and power supply. The rise time of this amplifier is about 0.15 microsecond, and the gain is about 300,000. In order to see the shape and beginning of light pulses, a Sickles delay line has been employed with delay time of one microsecond and rise time about 0.25 microsecond.***

The Model 501 amplifier has two equivalent cathode follower outputs. When counting pulses,

*** The technique employed is similar to that described in an article by W. C. Elmore, Nucleonics **2**, 16 (March 1948).

one output was brought to the scaling unit through a "T" fitting and a coaxial cable. Another cable led from the "T" fitting and was brought to an oscilloscope for routine examination of pulses while counting. The second cathode follower output was not used when such counting was studied. When the photographic studies of the rise time of pulses were made, the cable leading to the scaling unit was removed, and the short cable to the oscilloscope was the only output connection of the amplifier. The photographic studies of pulse shapes were made with the "laboratory oscilloscope" of the type described by Fitch and Titteron.[22] In other experiments, the second output was brought to a slave-sweep circuit (Model "260," designed by M. Sands) which permitted the use of a conventional oscilloscope for pulse observation.

The pulses were counted by a discriminator-scaling unit designed by M. Sands. This unit, called the "ten-channel discriminator," has ten channels into which all the incoming pulses from the amplifier are sorted. The pulses are counted in channels according to their height. For example, when 5-volt channels are used. channel 1 selects pulses from 5 to 10 volts. Channel 2 selects pulses from 10–15 volts, and so on. Channel 10 is a "surplus" channel and registers all pulses not already recorded by the other nine channels. A totalizer is available which counts all the pulses received. A cross check is therefore available to see that counts have not been missed. The totalizer is a scale of 32, the 9 channels are scales of 8, and the surplus channel is a scale of 16. A "sliding pulser," designed by M. Sands and W. A. Higinbotham, was also used to check the voltage width of the channels.

B. Preparation of the Phosphors

Most of the NaI(Tl) samples are prepared in the following way. Ten grams of NaI reagent (Baker and Adamson) are funneled into a half-inch quartz tube (closed at the lower end) to a depth of two or three inches. A constriction in the quartz tube, above the level of the NaI powder, permits subsequent sealing off of the sample while under vacuum. The quartz tube is connected to a "Hyvac" pump through a length of rubber hose and evacuated. The powder shows a tendency to "bump" and may travel rapidly throughout the vacuum system. To prevent this, a pad of glass wool is placed between the constriction and the end of the quartz tube, adjacent to the rubber hose. After evacuation, the quartz tube and contents are heated by a gas-air flame until the powder melts at about 650°C, at which the liquid NaI is pale yellow. The flame is then withdrawn and the melt allowed to cool directly. During cooling, the NaI crystals are formed and may be one or a few millimeters on a side. These crystals may be faintly luminescent. A discharge in the quartz tube, produced by the conventional "leak tester," is a good indicator of the luminescence of the phosphor. The tube is now taken away from the hose, opened to air, and 100 milligrams of Tl dropped into the tube. It is replaced on the vacuum system after the glass wool has been removed and discarded. The quartz tube and contents are reheated to melt the NaI and to mix the TlI with the NaI. After cooling and crystallization of the mixture, bright blue luminescence of the crystals can be observed with the leak tester discharge. The tube may now be sealed off for use.

Useful samples have also been made by mixing the two powders and employing a single heating process. It is probable that this method is equivalent to that described above. However, the most successful specimens were made by the process involving two meltings.

A good specimen is not always produced. The reason for this is not understood, although it is suspected that impurities, overheating, or too rapid heating before water vapor is removed may be responsible for the poorer specimens. The

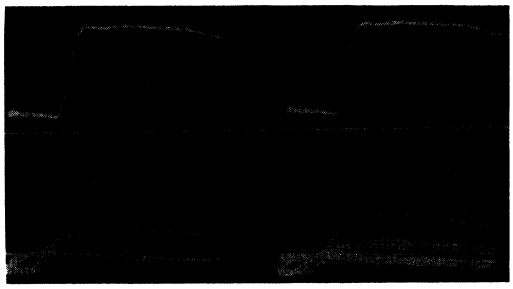

FIG. 3. "Pulser" pulses (above) and NaI(Tl) pulses (below).

[22] V. L. Fitch and E. W. Titterton, Rev. Sci. Inst. 18, 821 (1947).

poorer specimens are generally gray while the best specimens are clearer and white.

Single crystals have been prepared with surprising ease by taking the tubes as described, but with conical lower ends, suspending them in a furnace by a hook and wire, and then lowering slowly with a gear train and motor. This method of crystal formation, originally proposed by Bridgman, usually produces several large crystals. Such crystals are extremely efficient phosphors. For example, one specimen, containing about 4 grams, was made in the form of a few large crystals, one being quite large, of the order of a centimeter on one side. Even though the quartz tube was clouded by the heating process and the crystal was oblique to the walls, this specimen produced almost as many counts and nearly the same light output as a mass of 10 grams of small crystals when each tube was exposed to radium gamma-rays under the same conditions. Larger single crystal samples have been successfully grown in one-inch quartz tubes. The inner surface of such tubes apparently becomes etched in the process, and the output of light from these specimens is probably not as high as one might obtain if the surface remained clear. Large crystal specimens have been removed from the quartz tubes but rapidly "frost up" and become yellow in a humid atmosphere. Undoubtedly more successful results will be obtained when methods are developed for protecting the crystal surfaces from water vapor.

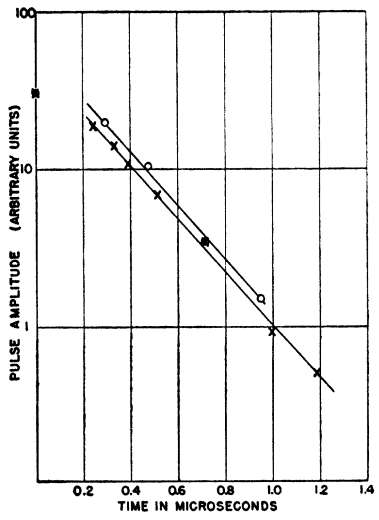

FIG. 4. Decay curves for NaI(Tl).

Figure 2 shows the active end of the first prepared phosphor tube of polycrystalline NaI(Tl). Examination of tube contents shows that the clear crystal region extends about two or three millimeters from the inside wall. Inside this region the material is more like coarse granular powder. Shorter specimens of the polycrystalline material have been prepared in tubes which, when sealed off, are 2.75 inches long.

Rough experiments have been made to find the most desirable amount of impurity. Specimens made by adding one percent of TlI seem to give the best results. Samples made with 200 mg, 50 mg, and 20 mg of TlI to 10 grams of NaI do not seem to provide as much light output as those made with 100 mg of TlI, although active samples have been prepared with as little as 0.02 percent TlI. However, the method by which these samples were made does not insure that the actual impurity content of the crystals is that determined by the amount added. The formation of gray crystals is different in different specimens and may also influence the reported findings. It has been found, however, that the single crystals are always "water white" and clear, thus indicating that the crystal is most satisfactorily prepared in a furnace under controlled temperature conditions. The torching method is probably not more than an expedient to obtain samples quickly. Counting samples have also been made with TlCl and TlBr.

IV. RESULTS

A. Counting Studies

In this section some data have been collected concerning the use of NaI(Tl) as a material in a scintillation counter. Such data may also be of interest for the theory of ionic crystals, since the alkali halide-thallium phosphors are simple fluorescent systems. Interesting topics are: (1) The duration of the light emission process. (2) The amount of light emitted per incident energy of ionizing radiation. This amount may vary for different particles of the same energy. (3) The sensitivity of the alkali halide scintillation counter in distinguishing different monoenergetic gamma-rays or beta-particles. (4) The spectral distribution of the emitted light. (5) The sensitivity to different radiations, e.g., alpha-particles, protons, neutrons, mesons, etc. (6) Mis-

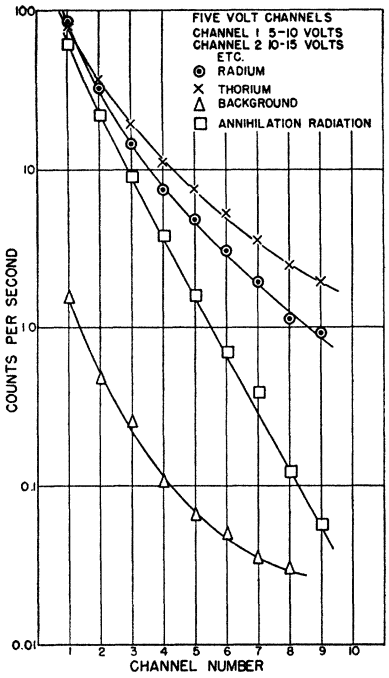

FIG. 5. Differential bias curves showing energy discrimination for various radioactive sources.

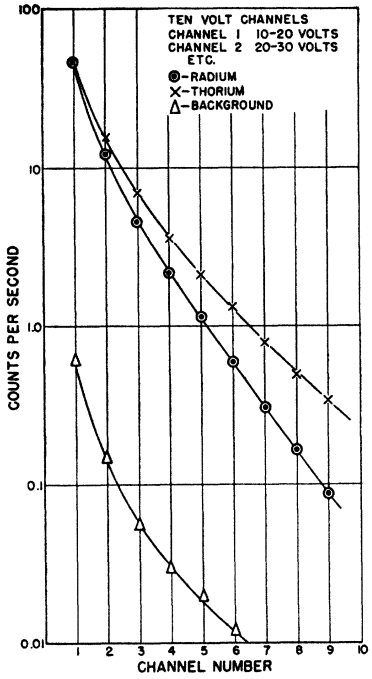

FIG. 6. Differential bias curves with ten-volt channels for radium and thorium sources.

cellaneous tests designed to see whether the alkali halide scintillation counter can serve as a general gamma-ray detector. (7) The "counting efficiency" of the crystal material, i.e., the ratio of the number of counted events to the ionizing flux passing through the crystal material.

It is not possible to present at this time complete information on all these points or on other interesting topics such as temperature effects.† However, a certain amount of information is available on almost all the topics mentioned. The points will be taken up in the above order.

(1) The emission of light, following the ionization produced in the NaI(Tl) phosphor by a gamma-ray encounter with the phosphor, has been studied by photographing the rise of the voltage pulse appearing across the anode load resistor of the photo-multiplier. In this study the anode resistor had a value of 10 megohms. The "clipping" or differentiating time constant of the amplifier was set at 40 microseconds. With this long clipping time, the height of the voltage pulse is proportional to the integral of the light emission curve and, therefore, represents the decay curve of the phosphor. This statement is true only if the phosphor decay time is several times as long as the amplifier-delay line rise time. For these experiments this is, unfortunately, not the case. Therefore, the quantitative studies must be considered, for the present, only as approximate.

To observe the effect of the amplifier-delay rise time, the phosphor pulses were compared with "artificial" repetitive pulses produced by a "Model 100" pulsing circuit. This "pulser" manufactures a step pulse with a rise time considerably smaller than that of the amplifier-delay line combination used in these experiments. Therefore, a pulser voltage pulse shows directly the rise time of amplifier and delay line. Such pulses are shown at the top of Fig. 3. Directly below are shown pulses due to gamma-rays striking the phosphor, with amplifier-delay line characteristics unchanged. The comparison between these photographs permits an approximate estimate of the decay time of the phosphor, as indicated below.

Figure 4 shows two plots which were obtained from the photographs of Fig. 3. The phosphor curves of the two largest pulses were normalized

† W. C. Elmore and R. Hofstadter, Phys. Rev. **75**, 203 (1949).

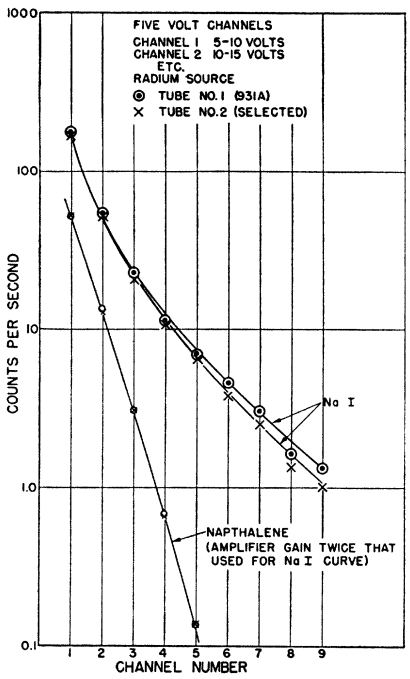

FIG. 7. Behavior obtained with different photo-multiplier tubes: amplifier rise time 0.25 microsecond; clipping time about two microseconds.

to the same maximum value. The differences between points on such normalized curves and the final saturation values of the pulses are the values plotted in the curves of Fig. 4. The figure shows that, except for the first points, both curves are fitted rather well by straight lines on semilog plots. When determined in this way, the time constants of the decays are 0.25 and 0.26 ± 0.05 microsecond. Since the 10–90 percent rise time of the "pulser" pulses is 0.27 microsecond, it can be seen that the values obtained for the phosphor will be only approximate. The phosphor measurements are probably correct to within 25 percent. The light pulse in this NaI(Tl) phosphor sample has a total duration, therefore, of the order of 0.8 microsecond. Although the decays studied appear to be exponential, other types of decay are not ruled out, particularly since only the larger pulses have been available for study.

In the experiments just described the voltage pulses measured were about 1.6 and 2.0 millivolts at the anode of the photo-multiplier.††

(2) The photo-multiplier pulse corresponding to a single ionization event depends on at least: (a) the energy lost in the crystal by the beta-particle released by the gamma-ray, (b) the region in the polycrystalline sample in which the beta-particle is released, and (c) the statistical secondary multiplication in the dynodes of the photo-multiplier. Item (b) is important since it includes the light path, number of reflections and transmissions at boundaries, and the absorption of light within the sample. The amount of light received by the photo-multiplier is, of course, further determined by the optical arrangement used to direct light from the polycrystalline sample into the photosensitive cathode of the photo-multiplier. It may be expected that with very large light pulses the voltage pulses will be proportional to the amount of light received by the photosensitive surface.†††

A very rough figure for the amount of light emitted in one of the large scintillations due to radium gamma-rays may be obtained by comparing the voltage pulses due to gamma-rays and those due to a stimulated phosphorescence in the crystal. Weak phosphorescence may be excited in the phosphor by exposing the phosphor tube to a strong ultraviolet source or by producing a discharge in it with a "leak tester." It may be expected that for weak phosphorescence indi-

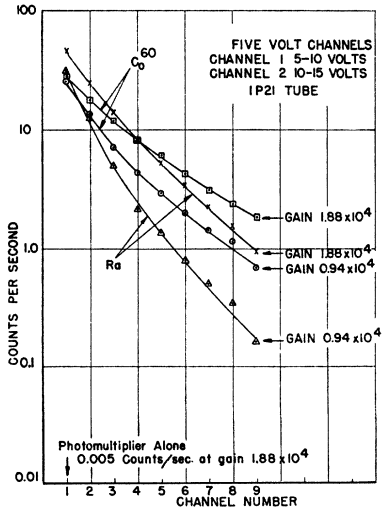

FIG. 8. Counting rates at different amplifier gain settings.

†† Larger pulses are easily obtained by increasing the photo-multiplier voltage. The pulses used are of convenient size in working with the 501 amplifier.

††† At the Rochester conference on high speed counters, Miss Rosalie C. Hoyt presented calculations on such matters.

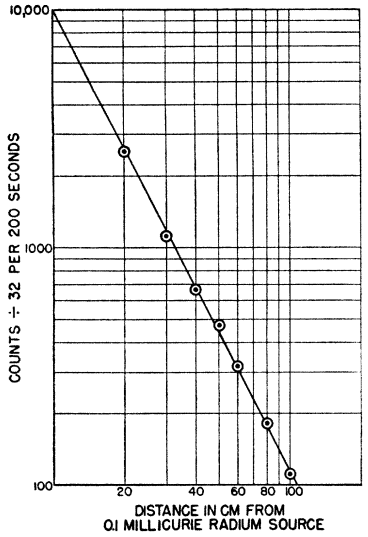

FIG. 9. Test of inverse square law.

vidual photo-multiplier counts will correspond to single electrons emitted at the photo-cathode.

Some of the large pulses due to gamma-rays are of the order of twenty-five or thirty times the height of pulses observed in phosphorescence, when the comparison is made visually on an oscilloscope screen. Since the efficiency of the photosensitive surface of the photo-multiplier lies between two and five percent, the number of photons incident on the photosensitive surface is of the order of one thousand. We may take five or ten percent as a rough estimate of the fraction of light collected by the photo-multiplier to that emitted by the crystal sample. This figure is obtained by estimating the relative aperture. We then obtain a probable figure of from 10,000 to 20,000 photons emitted by NaI(Tl) polycrystals due to an energetic gamma-ray from radium. This figure is probably a lower limit for the number of photons in such a large scintillation. Nevertheless, this figure differs considerably from the value 200,000 photons given by Moon[6] for materials which show smaller light efficiencies than NaI(Tl). It would be desirable to make more accurate determinations of the light yields from such phosphors.

(3) Deutsch[23] has shown that gamma-ray energies can be distinguished in naphthalene counters by means of bias curves, or, in other words, by the distribution pattern of pulse heights. We have shown that energy discrimination is possible with NaI(Tl) scintillations, although care must be exercised in the interpretation of the bias curves. Figure 5 shows differential bias curves taken with the ten-channel discriminator for radium, thorium (oxide), and C^{11} annihilation radiation sources. These curves were taken with channels five volts wide. Figure 6 shows curves for radium and thorium (oxide) with ten-volt channels. It can be seen that the graphs taken with the ten-volt channels show linear behavior at the higher channel numbers. It is also observed that the annihilation radiation graph is practically linear over its whole length. These facts suggest that the behavior in the higher channels may be linear (semilog plot) for homogeneous gamma-rays. The cobalt 60 curves of Fig. 8 are in reasonable agreement with this view.

It can be seen in Fig. 8 that the cobalt 60 curves show smaller slope than the radium curves in the region examined. This is probably connected with the fact that the cobalt radiation is almost homogeneous (1.2 Mev), while the

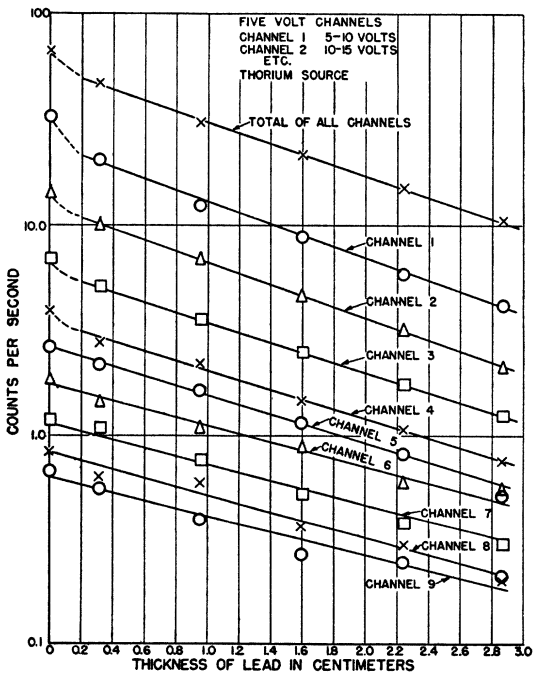

FIG. 10. Lead absorption curves taken with NaI(Tl) counter.

[23] M. Deutsch, Tech. Report No. 3, Laboratory for Nuclear Science and Engineering, M.I.T., December (1947).

TABLE I. Results of absorption measurements.

	Total	*Ch1*	*Ch2*	*Ch3*	*Ch4*	*Ch5*	*Ch6*	*Ch7*	*Ch8*	*Ch9*
Absorption coefficient (cm^{-1})	0.59	0.62	0.61	0.53	0.53	0.53	0.45	0.44	0.47	0.43

radium radiation is complex and has a strong, soft component.

Thus, while the rules of interpretation of these curves are not yet clear, it appears that the scintillation counter shows possibilities of energy discrimination in addition to its normal counting function.††††

(4) By placing a luminescent crystal or polycrystalline sample and radioactive source in front of the slit of a spectrograph, one may obtain a photograph of the spectrum of the light emitted by the sample under bombardment. Such work has been carried on by Mr. J. C. D. Milton and the author and will be reported separately. The results will merely be stated here. The light from NaI(Tl) samples, when exposed to radium gamma-rays, lies in the region 3000A to 4900A with maximum lying between 4050 and 4150A. The width at half-maximum is generally about 800A, and there is some evidence of asymmetry in regions of smaller wave-lengths. The emission band therefore has a considerable visible component. Unfortunately, the spectrum induced by ultraviolet light in NaI(Tl), with which this data can be compared, is unknown.

(5) Scintillations in NaI(Tl) caused by radium alpha-particles have already been reported to be large compared with gamma-ray scintillations, and equal in magnitude to those in ZnS(Ag).[9] There is nothing further to be added at this time. Experiments with other particles such as protons, neutrons, mesons, etc., have not been attempted in any detail.

(6) Various studies of the NaI(Tl) scintillation counter have been made with the aim of learning its general properties when used as a gamma-ray detector. Among the topics studied are: (a) Behavior of the counter when different photo-multiplier tubes are used. (b) Photo-multiplier noise background. (c) Comparison of single crystal bias curves with polycrystal bias curves. (d) Number of counts as a function of source distance. (e) Use of scintillation counter with gamma-ray filters. (f) Background with NaI(Tl) counter. (g) Comparison of NaI(Tl) with anthracene and naphthalene.

(a) In these tests a NaI(Tl) phosphor tube was used both with a conventional 931A type and with a selected 931A photo-multiplier. The latter tube is the one used in most (but not all) of the experiments described in this paper. A comparison of the bias curves shown in Fig. 7 shows that the counting behavior is identical within experimental error for these two tubes. The only difference between the two cases rests in the operating voltages. The selected tube required 432 volts and the other tube 478 volts to obtain

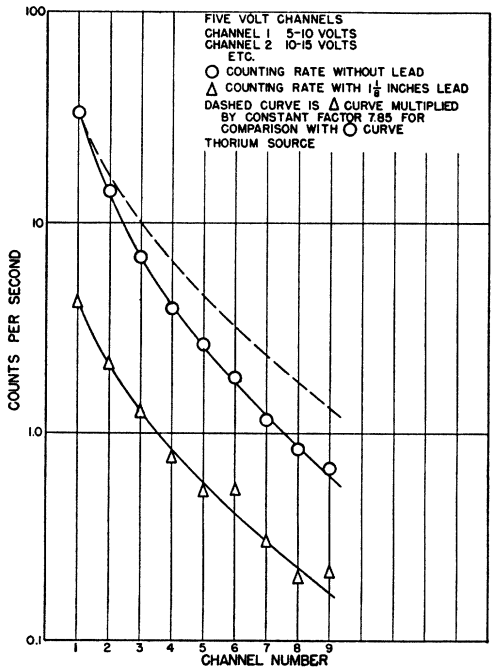

FIG. 11. Comparison of filtered with unfiltered radiation.

†††† *Note added in proof:* Hofstadter, Milton, and McIntyre (Bull. Am. Phys. Soc., No. 1, Jan. 1949, p. 16, E11 and in an article to be published) by using single crystals of NaI(Tl), cemented to the photo-multiplier tube, and a collimated Co^{60} gamma-ray source, have shown: (a) while the polycrystalline curves show the behavior of Figs. 5, 6, 8, 14(a), the single crystal Co^{60} curves show definite maxima in pulse size distributions. This fact will permit improved energy discrimination. Similar work has been carried out by P. R. Bell with anthracene crystals and beta-particle sources. (b) A very close approach to a plateau in counting gamma-rays can be obtained. For example, a variation of photo-multiplier voltage of 80 in 720 volts resulted in a change of 9 percent in total counting rate. It has been estimated that 93 percent of all Co^{60} gamma-ray encounters with the crystal are counted. The newer curves, corresponding to Fig. 14(a) drop almost to zero at higher voltages.

the results of Fig. 7. Curves taken with naphthalene are also shown in Fig. 7. It is perhaps fortuitious that the comparison is as close as has been found, for it is probable that some unselected tubes have equal noise level and lower photosensitivity than the two tubes tested.

(b) The noise level of a 1P21 photo-multiplier tube, with which the data of Fig. 8 were obtained, is too small to show as a curve in this figure. The noise pulses occurred at the rate of 1 pulse per 200 seconds and appeared in the first channel, although an occasional pulse appeared in a higher channel. The bias chosen in these tests might correspond to average operating conditions.

(c) It has already been mentioned above that a small NaI(Tl) sample (~4 grams) in the form of a few large crystals gave results comparable with a larger sample (10 grams) of polycrystalline material. Most of the inside surface of the former "single" crystal tube was frosted in the process of preparation. One clear oval portion, on the wall of the quartz tube, was not attacked. The tube was therefore mounted so that the clear portion faced the sensitive surface of the photo-multiplier. The pulse distributions obtained with this sample and with a polycrystalline sample were examined and found to be identical for equal numbers of counted pulses.††††

(d) If the observed pulses are due to direct transmission of gamma-rays from the source to the crystal specimen, the number of pulses should fall off as the inverse square of the distance. Figure 9 shows the results of making such a test. The background counting rate was subtracted for all points. At the largest distance (100 cm) the background correction amounts to 16 percent. Conformity with the inverse square law is therefor obtained, implying that counting of scattered gamma-rays can at most be a small effect for the bias and photo-multiplier voltages used.

(e) Figures 10 and 11 show the results of tests performed by placing lead absorbers of different thicknesses in front of a thorium source. From Fig. 10 one can calculate absorption coefficients from the straight lines for the different channels. The results are given in Table I.

It may be seen that, although the accuracy is not high, the transition to higher channels corresponds to measuring harder components, a result to be expected. The average value of 0.45 cm^{-1} for the last four channels compares with a calculated value of 0.47 cm^{-1} for the energy 2.62 Mev, which is the well-known thorium line. Considering the accuracy of this experiment, the measured result is in satisfactory agreement with the calculated value.

The curves of channels 1 to 4 show deviations from straight lines for zero absorber thickness, indicating that softer components probably accompany the 2.62-Mev line.

Figure 11 shows part of the data of Fig. 10 plotted in a different manner. The curves of Fig. 11 are direct differential bias curves taken without absorber (upper solid curve) and with a 1.125-inch lead absorber present (lower solid curve). For comparison, the lower curve has been shifted upwards (dashed curve) so that the first channel points coincide. It is seen that the bias curves again point to energy discrimination as discussed above in (3). The dashed curve indicates a harder gamma-ray than the upper solid curve. If extended further to the right, it is possible that the two curves would become parallel.

(f) The lower curve of Fig. 12 shows a differential bias plot of the background counts observed with an 8.6-gram mass of NaI(Tl). From

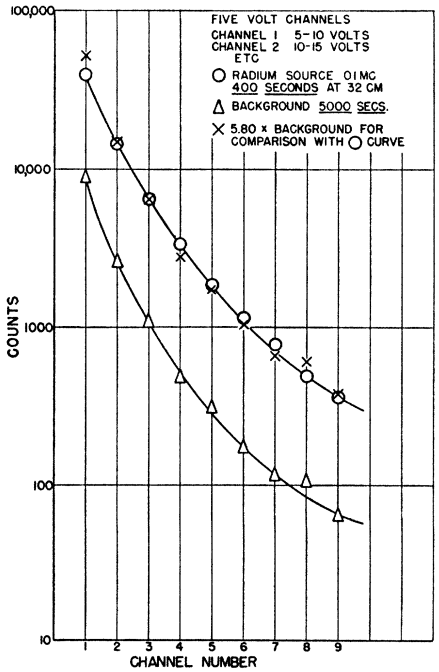

FIG. 12. Comparison of radium and background pulse distributions.

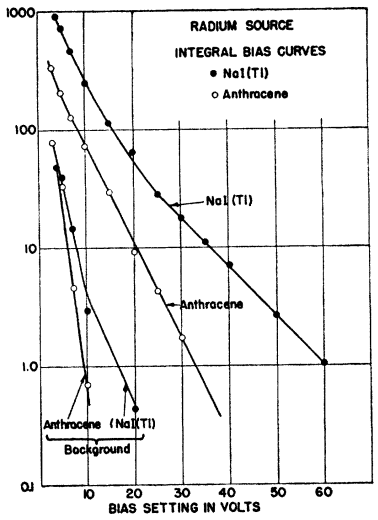

FIG. 13a. Integral bias curves for NaI(Tl) and anthracene for a radium source: amplifier rise time 0.8 microsecond; clipping time 0.8 microsecond.

this curve it can be seen that the total background counting rate is 2.8 counts per second. A comparison of this counting rate has been made with a Victoreen Geiger-Mueller type counter (diameter, $1\frac{1}{8}$-in., $2\frac{1}{2}$-in. length, $\frac{1}{32}$-in. Cu wall thickness). The G-M counter recorded pulses at the rate of 0.90 count per second.

An additional comparison of the counting rates of the NaI(Tl) counter and the G-M counter was made by exposing each to the same source at equal distances. Each counter was placed a distance of 33 centimeters from 0.1-millicurie radium source. The NaI(Tl) counter gave 168 counts/sec., while the G-M counter gave 32.5 counts/sec. Subtracting the background counting rates from each, the results are 165 and 31.6 counts/sec., respectively. The ratio of these counting rates is, therefore, 5.2. The operating conditions for the NaI(Tl) counter were 432 volts (selected 931A tube) and amplifier gain of 3.8×10^4. The bias was set so that channel 1 recorded pulses 5–10 volts, etc.

The counting rate of the NaI(Tl) counter for gamma-radiation is 5.2 times the counting rate of the G-M counter, despite the fact that the solid angle, subtended by the NaI(Tl) at the source, is only about one-third of that subtended by the G-M counter. One might, therefore, expect that the background counting rates should be in the same ratio, after correction is made for the counting of cosmic rays by the two counters. (Correction for contamination is too uncertain to be made.) The G-M counter may be corrected to read 0.63 count/sec., allowing 0.27 count/sec. for cosmic rays. The NaI(Tl) corrected background reading will be 2.71 counts/sec. The ratio of these counting rates is 4.3 instead of 5.2. Considering the rough estimates of the corrections and the neglect of other corrections, the agreement is thought to be satisfactory. One may therefore conclude that the background readings with the NaI(Tl) counter, at the bias used, have given an indication of the gamma-ray flux in the room consistent with measurements of a G-M

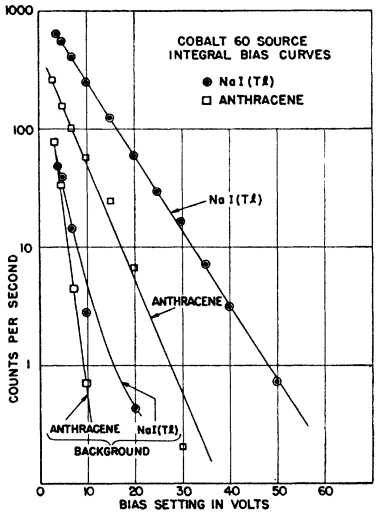

FIG. 13b. Integral bias curves for NaI(Tl) and anthracene for a Co^{60} source: amplifier rise time 0.8 microsecond; clipping time 0.8 microsecond.

counter, and therefore represent true gamma-background.

There are other indications that this conclusion is correct. These are as follows: (a) When two weak sources (0.1 millicurie each, normally kept 18 feet distant from the counter) were removed from the room, the NaI(Tl) background rate fell by 18 percent. (b) The distribution of pulse size closely resembles that of radium, which is suspected as the source of background counts in a physics laboratory. Figure 12 shows a comparison of the shape of the background curve and a curve due to a radium source. When the background curve is shifted upwards, the points of the lower curve fall rather closely on the upper

curve, due to radium. Furthermore, a closet about forty feet from the position of the NaI(Tl) counter is a storage point for radioactive sources, mostly radium, in Palmer Physical Laboratory. (c) Background counts taken with the NaI(Tl) and naphthalene counters showed the same ratio as the counting rates with a radium source present. This result may be seen in Fig. 2 of an earlier publication.[9]

(g) Integral bias curves comparing anthracene and NaI(Tl) have been taken with a Bell-Jordan amplifier and discriminator.‡ The rise and clipping times were selected as 0.8 microsecond. The sample of anthracene used was prepared by G. B. Collins and T. A. King. The anthracene sample weighed 2.0 grams and was clear except where cracked. The NaI(Tl) sample was of comparable size and polycrystalline. Figures 13(a) and (b) show the comparison. P. R. Bell previously reported similar findings at the Rochester conference on high speed counters. It should be recognized that both quantities, pulse magnitude and pulse duration, are of importance in comparing different phosphors. Anthracene and phenanthrene provide light pulses of initial height comparable with NaI(Tl) polycrystalline samples, but of smaller area when plotted against time. This behavior may be observed with a fast

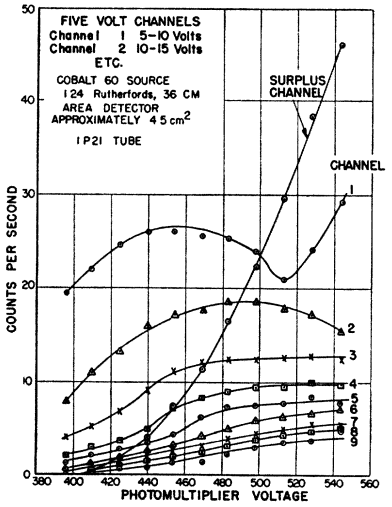

FIG. 14a. Counting rates in different channels as a function of photo-multiplier voltage for Co^{60} source. Background has been subtracted. The effective area of the counter is approximately 4.5 cm², and mass 13.0 grams.

‡ The author is indebted to Dr. H. Poss of Brookhaven National Laboratory for taking the data for these curves.

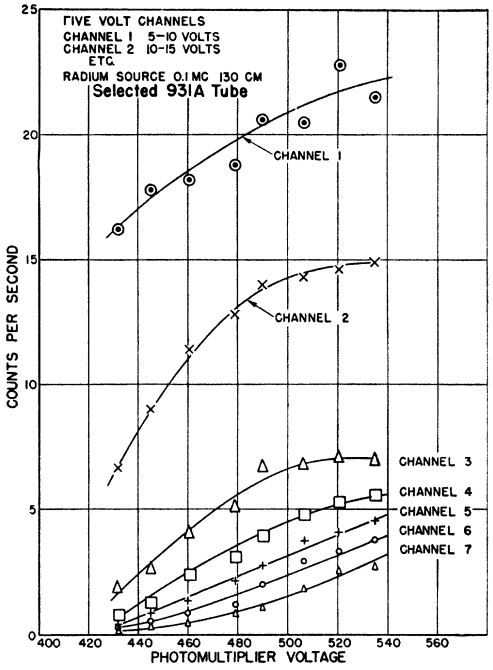

FIG. 14b. Counting rates in different channels as a function of photo-multiplier voltage for a radium source. Background has been subtracted. The effective area of the counter is approximately 3.6 cm², and mass 10 grams.

amplifier, of rise time 2×10^{-8} second.‡‡ With a slow amplifier the pulse height is proportional to the area under the light emission curves. Under these circumstances, NaI(Tl) pulses are much larger than anthracene (or phenanthrene) pulses. A single crystal of NaI(Tl) showed pulses about five times as large as a comparable anthracene sample with a slow amplifier. For fast counting, or low resolving times, phenanthrene and anthracene are definitely more suitable than NaI(Tl). A comparison of pulse size distributions for naphthalene and NaI(Tl) has been reported already.[9] There is nothing to add to these results at the present time.

(7) In this section the "counting efficiency" of the NaI(Tl) scintillation counter will be discussed. In experiments using counters a most important feature is that the counting rate should be independent, or nearly so, of counter conditions. In a Geiger-Mueller counter one has a "plateau" in counting over a certain voltage range. In this range the G-M counter is independent of other conditions if one excludes tem-

‡‡ To be described by W. C. Elmore.

perature variations and slow increase of products of chemical decomposition caused by long counting (10^9 counts or so). In the scintillation counter one encounters two variables which affect pulse magnitude, the voltage across the multiplier, and the gain of the amplifier. In the scintillation counter the pulses have variable magnitudes and, consequently, one always counts against an applied bias. In a G-M counter the pulses are of more or less uniform size, and the bias is unimportant.

In many respects the two variables, representing applied voltage and amplifier gain, are equivalent since both affect the amplification of the initial pulse. However, the noise of the photo-multiplier is changed by voltage, whereas amplifier gain, of course, does not influence the photo-multiplier noise but may bring it above the bias. (It is assumed, in this discussion, that amplifier noise is negligible.) It is possible, therefore, that different results will be obtained by separately varying photo-multiplier voltage and amplifier gain. In tests made of this point, both voltage and amplifier gain have been changed independently and the effects observed. As far as can be told, the two variables produce essentially the same results, so that the voltage region studied is that in which the noise does not increase more rapidly than the photo-multiplier gain increases as the voltage is raised. This result may be expected so long as the signal-to-noise ratio is high, as in these experiments. Figure 8 shows the results obtained when amplifier gain is changed.

The curves of Figs. 14(a) and (b) show the behavior of the NaI(Tl) scintillation counter with respect to variation of photo-multiplier voltage. These figures represent an effort to find a plateau in counting rate. It will be observed that an actual plateau has not been obtained so far, although the tendencies towards a plateau are quite clear, especially in Fig. 14(a).

It is to be expected that the counting rate will increase at low voltages because some pulses are still below the bias. At higher voltages the counting rates in the lower channels should decrease, while the higher channels, 3, 4, etc., should show evidence of saturation. With further increase of voltage the latter channels should show a decrease in counting rate. Evidence of this type is indicated by the curves of Fig. 14(a).

The second increase of counting rate in channel 1 of Fig. 14(a) is not understood, although it is quite possible that very low energy scattered gamma-rays from container, table, etc., and the overshoot of the amplifier for a large pulse may account for part of this increase. Curves 14(a) and (b) should be regarded as preliminary results. It is expected that further work will be performed employing a gamma-ray beam, a single crystal, and better shaping of the amplifier pulses. The single crystal should provide a higher signal-to-noise ratio. In Fig. 14(a) photo-multiplier noise sets in at about 520 volts, but has been subtracted as background.††††

The efficiency of the counter of Fig. 14(a) has been measured for Co^{60} gamma-rays by taking the total counting rate at 512 volts and dividing by the flux at the counter. The two figures are, respectively, 115 counts/sec. and 680 gamma-rays/sec. The measured efficiency is therefore about 17 percent. This figure is rather arbitrary since a plateau is not observed.

The efficiency for radium gamma-rays of the counter of Fig. 14(b) is more difficult to measure since the evidence of a plateau is less clear than in Fig. 14(a). It is also more difficult to find the true number of gamma-rays from radium, since there may be present some soft components difficult to measure. Using a value of 2.3 gamma-rays per disintegration of radium[24] and a counting rate of 93 counts/sec. at 538 volts (Fig. 14(b) and the totals of the remaining channels), one obtains an efficiency of about 65 percent. The effective area of the NaI(Tl) quartz tube is about 3.6 cm^2 in this case, and the 0.1-mc radium source is at a distance of 130 cm from the counter. This is a very high efficiency for the NaI(Tl), and exceeds an efficiency found by measuring the absorption of radium gamma-rays in an identical NaI(Tl) tube. The measured efficiency is 24 percent if it may be assumed that each absorbed quantum gives rise to one count. The observed counting rate is probably too high, although the figure of 2.3 gamma-rays per disintegration of radium may be too low.

Delayed pulses following alpha-particle scin-

[24] R. D. Evans and R. O. Evans, Rev. Mod. Phys. **20**, 305 (1948).

tillations have been observed by R. Sherr in ZnS counters. Such pulses were looked for in NaI(Tl), but were not found in a period of from two to ten microseconds following the main pulse. The high efficiency may be due to a longer-lived phosphorescence, although it is difficult to establish this point conclusively at the present time.

At the Rochester meeting on high speed counters, it was pointed out by several investigators that similar high efficiencies have been found in naphthalene and anthracene scintillation counters, and that the photo-multiplier itself may be producing delayed pulses. Further investigation of delayed pulses and efficiencies is clearly necessary.

B. Photographic Studies

The results of these brief studies are given at this point because of a possible application of the phosphor, photographic plate, or film technique to future experiments with gamma-rays or beta-particles and to radiation monitoring.

About a year ago the writer began a series of experiments to examine the luminescent light yield of single crystals under gamma-radiation. These experiments consisted of placing single crystal samples on top of a photographic plate. The light yield is then recorded by the photographic plate which acts as an integrating device. If a small portion of the crystal surface is masked by a piece of paper, or, better, if a metallic reflecting film is sputtered or evaporated on a portion of the crystal, the light output can be found directly as the difference between the blackening on parts of the photographic plate exposed to the light and those parts masked by the opaque film or paper. Similar early studies were made on CaF_2 by Winkelmann and Straubel,[25] with x-rays, shortly after the discovery of x-rays.

A single crystal of KI(Tl), previously described,[9] was placed on a photographic plate (Eastman 103-0) at a distance of one meter from a 1.8-millicurie radium source. A tube of NaI(Tl) was also placed on the plate. After thirty minutes of exposure, blackening under the luminescent samples was observed. The blackening under the NaI(Tl) tube was only slightly greater than that under the single crystal of KI. It appears, therefore, that the light output of KI(Tl) and NaI(Tl) are comparable, although the NaI(Tl) samples was one of only average sensitivity. The KI(Tl) sample was also favored by closer proximity to the plate.

It had previously been determined‡‡‡ that the KI(Tl) sample produced an increase of blackening, due to gamma-ray exposure, by a factor of one hundred, over the blackening resulting from direct exposure of the plate to the same radiation. Since NaI(Tl) samples have been made with twice the light output of the sample under test and since photographic film and a reflector could be wrapped around the tube for improved light reception, it seems reasonable that phosphor-film combinations could provide a factor of about four hundred in increased sensitivity of film to gamma-rays. Scattering experiments with gamma-rays might be attempted with a number of phosphor tubes placed at various angles around the scatterer.

Other tests, made with NaCl, KBr, and LiF powders to which thallium impurities were added, showed that NaI and KI powders were much more efficient as phosphors than the others.

It is interesting that KI(Tl) does not give large pulses with a photo-multiplier even though its total light output is comparable in magnitude and spectral distribution with that of NaI(Tl). One may draw the conclusion that light is emitted from KI(Tl) over a longer period of time than the light from NaI(Tl). The amplifier used in these tests had a rise time of 0.25 microsecond. It is possible that other KI(Tl) samples may behave differently; only two specimens of KI(Tl) have been studied.

ACKNOWLEDGMENTS

The author wishes to thank Mr. L. B. Harris for preparation of the quartz tubes, Mr. James Black and Mr. Howard E. Schrader for help with the drawings and photographs, and Mr. J. C. D. Milton for assistance with the equipment used in these experiments. Thanks are due Dr. P. C.

[25] A. Winkelmann and R. Straubel, Ann. d. Physik 59, 324 (1896).

‡‡‡ Tests were made of time of exposure *versus* blackening.

Gugelot for preparation of a C^{11} sample. Kind interest in these experiments by Drs. J. A. Wheeler, M. G. White, and M. ten Bosch is gratefully acknowledged. The loan of Navy project equipment (Contract N6ori-105, Task Order I) and of equipment by Drs. R. Sherr and H. W. Fulbright is also appreciated. The author is further indebted to Mr. T. A. King for a sample of anthracene and to Dr. H. Poss for study of this specimen. Thanks are due the Office of Naval Research, the University of Rochester, and conference members, for the exchange of ideas at the recent Rochester conference on high speed counters. Finally, the author wishes to thank the Brookhaven National Laboratory for an opportunity to continue part of this work on counters.

PHYSICAL REVIEW VOLUME 75, NUMBER 5 MARCH 1, 1949

Further Experiments on Cosmic-Ray Bursts*

H. S. BRIDGE AND B. ROSSI
Physics Department and Laboratory for Nuclear Science and Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts

(Received November 29, 1948)

The paper describes further results obtained by the observation of coincidences between pulses of a Geiger-Mueller counter tray and of an ionization chamber. The experiments were carried out partly aboard a B-29 aircraft and partly on the top of Mt. Evans in Colorado. The main purpose of the experiments at Mt. Evans was an investigation of the "transition curve" in lead. The experimental data are consistent with the assumption that the coincident bursts observed with a lead shield between the Geiger-Mueller tubes and the ionization chamber are mainly produced by cascade showers initiated by electrons and photons either incident upon the lead from the atmosphere or produced in the lead by nuclear interactions. The experiments aboard the B-29 furnish information on the variation with altitude of the radiation responsible for the nuclear interactions.

I. INTRODUCTION

EXPERIMENTS made by means of a tray of Geiger-Mueller counters and an ionization chamber, arranged one above the other and separated by several inches of lead, revealed the existence of time coincident pulses, whose rate of occurrence increases rapidly with altitude.[1] This phenomenon was interpreted by postulating the existence of ionizing particles different from electrons or ordinary mesons, which are much more abundant at high altitude than at sea level and are capable of producing secondary electrons or photons of high energy after traversing moderate thicknesses of lead. The electrons or photons undergo cascade multiplication in the lead, and the resulting showers are responsible for the ionization bursts in the chamber.

In order to test this interpretation, an experiment was carried out[2] in which part of the solid lead absorber between the Geiger-Mueller tray and the ionization chamber was replaced with a cloud chamber containing a number of lead plates; the expansion was triggered by the coincident pulses of the Geiger-Mueller tray and the ionization chamber. The cloud-chamber pictures thus obtained showed that shower production by penetrating particles was indeed responsible for a large fraction of the coincidences. Furthermore, they showed that the showers usually contained electrons as well as penetrating or heavily ionizing particles. This was taken as evidence that the showers originated in nuclear, rather than in electromagnetic, interactions. The

* The work described in this paper was assisted by the joint program of the Office of Naval Research and the Atomic Energy Commission. The B-29 aircraft was provided by the U. S. Air Forces. The facilities for the work at Mt. Evans were supplied by the Inter-University High Altitude Laboratory.

[1] H. Bridge, B. Rossi, and R. W. Williams, Phys. Rev. **72**, 257 (1947).

[2] H. S. Bridge, W. E. Hazen, and B. Rossi, Phys. Rev. **73**, 179 (1948); H. S. Bridge and W. E. Hazen, Phys. Rev. **74**, 579 (1948).

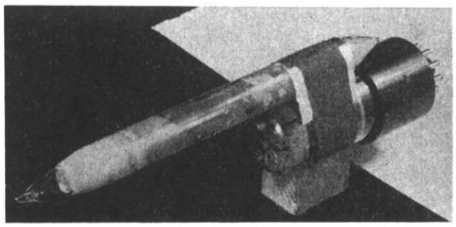

FIG. 1. Phosphor tube mounted on photo-multiplier.

FIG. 2. Active region of tube of Fig. 1.

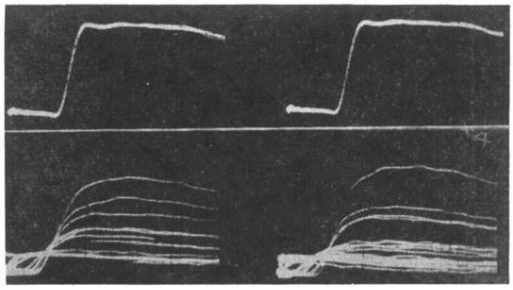

FIG. 3. "Pulser" pulses (above) and NaI(Tl) pulses (below).