THE EFFECT OF TEMPERATURE ON THE PERFORMANCE
OF A NaI-Tl SCINTILLATION SPECTROMETER

by

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A THESIS
SUBMITTED TO THE FACULTY
IN PARTIAL FULFILLMENT OF THE
REQUIREMENTS FOR THE DEGREE OF
MASTER OF ARTS

Houston, Texas
June, 1951
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INTRODUCTION

It has been found recently that a transparent phosphor in combination with a photomultiplier tube makes a gamma ray spectrometer of low resolution and high gamma ray conversion efficiency. A gamma ray photon produces a light pulse, or scintillation, in the phosphor. This in turn appears at the output of the photomultiplier as a pulse of electrons, which is converted into a voltage pulse by an amplifier.

While some liquid phosphors have been successfully used\(^1,2\), our work to date has been done with crystal phosphors, one of the best of which is NaI, activated with 0.5% Thallium\(^3,4\). This crystal has been used, in spite of its hygroscopic nature, because it produces large scintillation pulses, having a decay time of 0.254-sec.\(^5\) and because it has a high efficiency for absorbing gamma rays.

DISCUSSION OF NaI-Tl PHOSPHOR

Whether the gamma ray is absorbed by photoelectric, Compton, or pair production process, a high velocity electron results, which, in traversing the phosphor, loses its energy in excitation and ionization.
In returning to their ground state, these atoms of the phosphor radiate this energy in the form of light through the crystal to the photomultiplier tube.

Not all of the incident energy is converted into light photons, however, and Kallman\(^6\) denotes as the **physical efficiency** of a phosphor that fraction of the energy lost by the gamma ray which is converted into light energy. If, for example, some 50 ev of gamma ray energy are expended per photon of wavelength 4800 Angstroms in the NaI-Tl phosphor, the physical efficiency would be approximately 5\%. Of course, all the radiated light may not reach the photocathode surface. ZnS, for example, absorbs its own radiation and is opaque at thicknesses greater than 80 mg/cm\(^2\).\(^{10}\) Kallman\(^6\) denotes as the **technical efficiency** that fraction of the initial energy which finally reaches the photocathode surface. For NaI-Tl, the technical efficiency is high because this crystal is transparent to its own radiation. This allows the use of large crystals, which have the advantage of absorbing higher energy gamma radiation, and also of absorbing both the X-rays from ionized atoms and the scattered Compton photons.

The spectrum taken with the NaI-Tl scintillation spectrometer shows two or three or sometimes more peaks for each gamma ray. This makes analysis,
particularly of a complex gamma ray distribution, somewhat more difficult, but it can be done if care is taken to make sure of the identity of the peaks. The photopeak appears at the energy of the gamma ray, because the X-ray emitted by the ionized atoms has a very high probability of being completely absorbed in the crystal. If, however, some scattered gamma rays escape without further interaction, a Compton distribution will be formed, with its upper edge at the maximum energy of the Compton recoil, viz. \( \frac{2E_y^2}{E_0} \), where \( E_y \) is the energy of the incident gamma ray and \( E_0 \) is the rest energy of the electron. Pair production in the phosphor will give pulses at the energy of the incident radiation if both photons from the annihilation of the positron are absorbed in the crystal. If, in an appreciable fraction of the cases, both escape, a peak is formed 1.02 Mev below the photopeak, and if, in a sufficiently large percentage of the events, one escapes, a peak may show at 0.51 Mev below the photopeak.

Figure 1 shows the cross sections in NaI-Tl for the three processes as a function of gamma ray energy. This figure shows clearly how the absorption process varies from being principally photoelectric at low energy to principally Compton at medium and pair absorption at higher energies.
From these curves, one may make an estimate of the efficiency of absorption in a NaI-Tl crystal. The total linear absorption coefficient for 1 Mev gamma rays, calculated from the curves of the cross sections, is 0.215 cm$^{-1}$. This represents an efficiency of 41.5% for absorbing gamma rays traversing one inch of crystal. Using approximate range-energy relations for electrons in matter, the range of a 1 Mev electron is 0.1 cm in NaI-Tl. Taking account of this factor, which shortens the effective thickness of the crystal for absorbing gamma rays, decreases the efficiency by $1\frac{1}{2}$%.

For a 0.5 Mev gamma ray, the efficiency is calculated to be approximately 62%. The range of the electron in the crystal is a negligible factor here.

The efficiency of NaI-Tl in absorbing a 3 Mev gamma ray is 23.3%, where the range of a 3 Mev electron (approximately 0.4 cm) has been taken into account. It should be noted that the minimum efficiency occurs at about 4.3 Mev and, still for a one-inch crystal, equals 21.3%.

We should note also that these figures do not predict the intensity of the peak at the gamma ray energy, since it is not possible to calculate either what fraction of the scattered Compton photons or what fraction of the annihilation photons are absorbed by the crystal.
STATISTICS

If a given energy gamma ray produced exactly the same pulse height every time, the peaks would be sharp lines. There is, however, a spread caused by various statistical factors. The energy expended per photon produced in the crystal is what determines the physical efficiency. In NaI-Tl, this averages about 50 ev per photon. If now a 0.5 Mev gamma ray is absorbed in the phosphor, it will produce some 10,000 photons. This number itself is subject to some statistical fluctuations, possibly of the order of \((10,000)^{\frac{1}{2}}\). In this case, since the crystal is transparent, almost all these photons will reach the photosurface of the tube. There, using an approximate value of \(0.5 \times 10^{-1}\) photoelectrons produced per photon, one would expect 500 photoelectrons. The root mean square deviation for these photoelectrons is \((500)^{\frac{1}{2}} = 23\), or approximately 5%. This is the fluctuation in the original number of photoelectrons. The question now becomes how much will the photomultiplier tube increase this fluctuation due to the fact that the secondary electrons produced at each dynode vary in a statistical manner, so that all pulses obtained from single photoelectrons are not the same height.
The statistical spread in output pulse will be the sum of the contributions of several causes, chief of which are: a) fluctuations in the multiplication factor of the tube, b) fluctuations in the number of photoelectrons emitted from the photocathode for a given quantity of light reaching it, and c) fluctuations in the physical and technical efficiencies of the crystal.

The larger the number of photoelectrons emitted from the photocathode surface, the smaller will be the relative effect due to cause a). Likewise, the larger the number of quanta emitted in the phosphor, the smaller will be the relative effects due to b) and c), and, of course, to a) also. It is of interest then, to find the average number of photoelectrons necessary to make the contribution due to a) negligibly small.

If \( X = \) multiplier gain, \( Y = \) number of photoelectrons produced by an incident particle (this will include effects b) and c) together), \( Z = \) number of output electrons produced by that incident particle, then

\[
Z = XY
\]

where \( Z \), \( Y \), and \( X \) are average values. H. S. Snyder of Brookhaven National Laboratory, in his analysis of the statistical problem of the photomultiplier tube, has shown, assuming no specific shape for the distributions,
that,
\[ \langle Z(z-1) \rangle_{av} = \langle Y(Y-1) \rangle_{av} X^2 + Y \langle X(X-1) \rangle_{av} \]  
(2)

From this, it can be shown that
\[ \frac{\langle z^2 \rangle - \bar{z}^2}{\bar{z}^2} = \frac{1}{Y} \frac{\langle x^2 \rangle - \bar{x}^2}{\bar{x}^2} + \frac{\langle y^2 \rangle - \bar{y}^2}{\bar{y}^2} \]  
(3)

or
\[ \sigma_z^2 = \frac{1}{\bar{y}} \sigma_x^2 + \sigma_y^2 \]  
(4)

where \( \sigma_z^2 \) etc. are the fractional mean square deviations of the distributions.

Clearly, the smaller the value of \( \bar{y} \), the greater is the contribution of the 1st term in equation (4), i.e., the fluctuations in multiplier gain, to the overall distribution. Of course, we must remember that for smaller \( \bar{y} \), the relative effects due to b) and c) will also increase somewhat.

Measurements were made by Hoyt\(^{13}\) with values of \( \bar{y} \) varying from 35 to 92 and values of \( \sigma_x^2 \) and \( \sigma_z^2 \) varying from 0.13 and 0.07 to 0.23 and 0.64 respectively. Her results indicate that, for \( \bar{y} > 25 \) the contribution due to fluctuations in multiplier gain is negligible. For example, if \( \sigma_z^2 = .300, \sigma_x^2 = .200 \), using average values indicated by Hoyt, and \( \bar{y} = 25 \), then \( \sigma_y^2 = .292 \), so that the contribution of \( \sigma_x^2 \) to \( \sigma_z^2 \) is very small compared to that of \( \sigma_y^2 \). Therefore the chief cause of spread is in the fluctuations of the efficiency of the photosurface and of the physical and technical efficiencies of the
crystal. This indicates the importance of having uniform phosphors of good physical efficiency.

PURPOSE OF PROJECT

The objective in this investigation was first to construct and put in working order a NaI-Tl scintillation spectrometer. When this was accomplished, it was felt worthwhile to measure the temperature dependence of the apparatus, which has been suspected of causing variation in performance. To our knowledge, this had not yet been attempted quantitatively in the region of room temperatures, in spite of its important bearing on the accuracy of data taken with this instrument. It was decided, therefore, to vary the temperature of the crystal over the extremes of room temperature, holding the temperature of the tube constant. Then, the tube temperature could be varied, holding that of the crystal constant.

R. J. Moon mentions some effects of cooling and heating on spurious pulses in both tubes and some crystals. Cooling CaF$_2$, for example, reduced spurious pulses and heating increased them. These spurious pulses probably have a considerable spread in energy and contribute a bit to each portion of the curve, tending to raise it slightly. Unless, however, there is
a concentration of these pulses in a region near the energy being investigated, and unless their counting rate is appreciable compared to that of the measured rays, this effect should not cause an appreciable shift in the position of the peak.

It is conceivable, perhaps, that an increase in heat motion might increase the rate of emptying of the metastable states in the crystal by collision. Thus an increase in temperature might cause a decrease in output pulse size. Also, heat motion might affect the emission properties of the photomultiplier.

EQUIPMENT

In order to achieve the objective of a working NaI-Tl crystal spectrometer, it was necessary to construct a photomultiplier tube mount and shield, a standard type preamplifier, a -150 volt regulated power supply adapted from a design by Elmore and Sands\textsuperscript{14}, and a single channel pulse height analyser of the type designed by Francis, Bell, and Gundlach\textsuperscript{15}. This last was by far the most intricate, but at the same time the most interesting and vital piece of electronic equipment used. A circuit diagram of it is shown in figure 2, and a block diagram of it is shown in figure 3.
DIFFERENTIAL PULSE HEIGHT ANALYSER

figure 2
Figure 3
PHOTOTUBE and PREAMPLIFIER CIRCUITS

figure 4
The general operation of the pulse height analyser is as follows: the 0 - 100 volt output pulse of a standard A-1 amplifier is fed into the pulse height selecting amplifier. This amplifier has a bias setting which can be varied through nearly the entire 0 - 100 volt range. If a pulse is large enough to pass the bias, this first section amplifies the 10 volt portion above the bias setting and puts it out as a 0 - 100 volt pulse into the upper and lower pulse height selector sections. The lower pulse height selector triggers on any pulse passing through, and transmits it to the anticoincidence circuit. The upper pulse height selector, however, also has a variable 0 - 100 volt bias setting. A pulse large enough to pass this bias will also reach the anticoincidence circuit, so that no output pulse will register from the analyser. If a pulse is large enough to pass through the amplifier section but not large enough to pass the upper pulse height selector, then an output pulse is transmitted by the anticoincidence circuit, recording the fact that a pulse within the height $\Delta E$ has entered the analyser. This $\Delta E$ range can be narrowed, with good results, to only a few tenths of a volt of the original A-1 pulse.

The advantages of this arrangement are obvious.
One gets a good differential curve directly from the data, improving considerably the statistics involved. Normally, the number of pulses above a value $E$ is counted for a fixed time $t_0$ to obtain the value $n_1$, after which the setting of the pulse height selector is changed to count all pulses above $E + \Delta E$ to obtain the value $n_2$. The number of pulses in the interval $\Delta E$ is $(n_1 - n_2)$ with a statistical error of $(n_1 + n_2)^{1/2}$. With this instrument, however, the number of pulses above $E$ and the number above $E + \Delta E$ are measured simultaneously and subtracted automatically. Thus the statistical error is immediately reduced to $(n_1 - n_2)^{1/2}$ and the counting time is greatly reduced for the same statistical error. In the integral method, moreover, dial setting errors are important in investigating a narrow $\Delta E$. Assuming the dial can be set accurately to 0.1% and a $\Delta E$ of 1% is used, the error of $\Delta E$ can be as large as 20%. The differential pulse height analyser thus offers a clear advantage because just one setting is needed. In addition, it makes it possible effectively to maintain a gate of, say, 0.5 volts of the original pulse spectrum by holding only a 5 volt bias in the upper pulse height selector. Clearly, then, the accuracy involved in holding a bias at .5 volt above $E$ by the former method, is here determined by the greater accuracy in holding a 5 volt bias
in the upper pulse height selector.

One other point should be mentioned. The use of a narrow gate is particularly advantageous if one desires to investigate a pulse spectrum in the presence of considerable background radiation. Usually the radiation being investigated is concentrated in a relatively narrow energy region, while the background is spread out. In the proximity of the Van de Graaff generator, for example, there will be X-rays at the lower end of the energy scale and background gamma radiation at the upper end. Use of the narrow AE cuts out all pulses of energy below and above the energy region being investigated.

The actual performance of the finished spectrometer is shown in figure 5. It is a gamma ray spectrum of Cs$^{137}$, taken using a 1x1x2 inch NaI-Tl crystal mounted on lucite, and the best of the three available 5819 photomultiplier tubes. The photopeak is shown at 433 divisions and represents the full energy of the 0.661 Mev gamma ray. The Compton distribution is not well resolved but can be discerned, breaking off at about 293 divisions, or 0.477 Mev. The low energy peak at 122 divisions, or 0.186 Mev, is due to back-scattered gamma rays from the aluminum cap. It is very near the minimum energy for a single Compton scattering (0.184 Mev).
PROCEDURE

To vary the temperature, it seemed simplest to surround the crystal and lucite light pipe with a coil of copper tubing. Water was circulated in this coil, and the temperature adjusted by means of a mixer type faucet connected to hot water and tap water outlets. The temperature was measured by means of a thermocouple inserted through the coil, between the coil and the light pipe. The thermocouple was attached to a sensitive suspension galvanometer which had been calibrated against a 0° - 60° C mercury thermometer and three checkpoints; melting ice, boiling, pure ethyl alcohol, and boiling distilled water.

In order to eliminate the effects of possible changes in the phototube itself, a control coil was wrapped around the outside of the mu-metal shield, and tap water circulated through this during the runs. The temperature of the tap water was quite constant, staying within one degree of 25° C during all the runs.

Since the hot water tap produced temperatures up to about 62° C, this arrangement was quite convenient for the high temperature measurements. For the low temperature measurements, tap water was first circulated through a coil immersed in a thermos jug filled with
ice and salt, before sending it through the coil around the crystal. In this case, the temperature was regulated by the rate of flow of water through the cooling coil, since the more time the water spent in the coil, the cooler it became. Hence a pinch clamp on the line leading to the cold water coil proved to be an efficient and quite sensitive temperature control.

The mu-metal shield around the tube was used to eliminate the effects of any stray magnetic fields, and the entire assembly was made light tight by a close-fitting cover sealed with black electrical tape. The experimental arrangement is indicated in figure 6.

In making the actual runs, a difficulty appeared which we were not at the time able to remove. The calibration runs made at room temperature, with tap water running through both coils, were not reproducible after about 10 hours within 3 to 4 divisions in 360, or 1%. For this reason, only temperature runs taken within 3 hours of a room temperature run were counted in analysing the data.

There also arose the question of how long it took the entire crystal to reach the hot or cold temperature being applied. For this reason, only data taken at least one hour after temperature was applied was counted as valid. These two criteria, unfortunately, reduced the amount of valid data to four runs.
figure 6
RESULTS AND CONCLUSIONS

The four runs used were taken at temperatures of 6°C, 14°C, and two at 43°C. The results are tabulated in table 1.

<table>
<thead>
<tr>
<th>Temperature (°C)</th>
<th>Peak Shift, divisions</th>
<th>Peak Shift, Mev</th>
<th>Peak Shift Percentage</th>
</tr>
</thead>
<tbody>
<tr>
<td>6</td>
<td>-7</td>
<td>-.013</td>
<td>-1.9%</td>
</tr>
<tr>
<td>14</td>
<td>-2</td>
<td>-.003</td>
<td>-0.51%</td>
</tr>
<tr>
<td>43</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>43</td>
<td>+2</td>
<td>+.004</td>
<td>+0.55%</td>
</tr>
</tbody>
</table>

These results are consistent, but determination of the peaks from curves drawn through the data points was not certain within 2 divisions, so that the reality of these indicated shifts is in doubt. At least 10,000 counts were taken at all points, and in the first and last runs of table 1, about 100,000 counts were taken at points around the peak.

The results, then, indicate that there is not any appreciable effect due to temperature variation of the crystal alone within the extreme ranges of what one might call room temperature. The next step would
seem to be that of maintaining the crystal at constant
temperature, while varying the temperature of the
photomultiplier tube, again allowing at least an hour
for the tube to reach equilibrium temperature.
ACKNOWLEDGMENT

The writer wishes to express his sincere appreciation to Dr. J. R. Rissel for his constant encouragement and guidance in carrying out this project; to Dr. W. H. Burke, Jr. for many hours of helpful advice and discussion; and to Mr. J. F. Van der Honst and the men of the physics shop for their invaluable aid and advice in the construction of equipment.
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