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The Electron Lifetime, an Experimental
Test of Charge Conservation

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1965

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THE ELECTRON LIFETIME,
AN EXPERIMENTAL TEST OF CHARGE CONSERVATION

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Doctor of Philosophy

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IN NUCLEAR SCIENCE ABSTRACTS

by

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ABSTRACT

New lower limits on the electron lifetime have been set by a series of experiments sensitive to two different conditions for non-charge-conserving electron decay processes. The lifetime of iodine K electrons for a process without photons among the decay products has been shown to be $\tau_e > 4 \times 10^{21}$ year compared to the earlier limit of $\tau_e > 10^{18}$ year set by der Mateosian and Goldhaber. The lower limit on the lifetime for the process $e^- \rightarrow \nu_e + \gamma$ has been raised from a previous best value of $\tau_e' > 10^{19}$ year to $\tau_e' > 5 \times 10^{22}$ year. These results have been interpreted in terms of their implications with respect to the law of charge conservation.

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I. INTRODUCTION

We are reminded on occasion as a consequence of experiments that the conservation laws of physics cannot be relied upon in regions where they have not been tested, which emphasizes once again the empirical nature of our science. One of these laws, the conservation of charge, is associated with the invariance of the total Lagrangian under certain gauge transformations and is generally taken to be absolute. However, regardless of how attractive these laws may appear theoretically, their final acceptance must rest on experimental evidence.¹ The downfall of parity conservation points up the danger in the application of conservation laws to unexplored phenomena.

The question of conservation laws and their experimental basis has been discussed in detail by Feinberg and Goldhaber.² In their discussion of an experimental test of charge conservation they point out that "...a particularly good place to look for...violations (of charge conservation) is to examine systems which cannot make any transitions at all if the conservation laws are satisfied, but could if charge were not conserved..." The electron meets this criterion because it is prevented from decaying to lighter particles only by charge conservation: all known particles lighter than the electron have charge zero. The lifetime of the electron is, therefore, a measure of charge conservation.

If we remove the constraint imposed by charge conservation, the electron is free to decay into a photon and a neutrino, a neutrino and two antineutrinos, or other combinations of photons, neutrinos and antineutrinos which conserve energy, momentum, spin, and lepton number. We are assuming here that charge conservation is the only law which is violated, although the simultaneous violation of two or more of the conservation laws is not inconceivable (for example as happened with parity and particle-anti-particle symmetry.)

The reaction $e^- \rightarrow \nu_e + \gamma$ is attractive to consider because both weak and electromagnetic interactions are represented, except for removal of the singularity associated with the charge. It is consistent with energy conservation since the neutrino and photon appear to have no rest mass. It is consistent with momentum conservation in that two or more decay products are sufficient to insure that the momentum remain zero in the center-of-mass system. Finally, the neutrino conserves the lepton number of +1 and the photon (spin 1) and neutrino (spin 1/2) combine to take up the spin (1/2) of the electron. It is interesting, though we do not take advantage of the fact in the present experiment, that the photon is polarized.

Similar arguments show the same consistency for the process $e^- \rightarrow 2\nu_e + \bar{\nu}_e$, and other sets of decay products can

be imagined such as $\nu_e + 2\gamma$, $3\nu_e + 2\bar{\nu}_e$, etc. We arbitrarily choose to concentrate our attention on the two-particle decay.

A particularly attractive experimental test of the electron lifetime has been carried out by der Mateosian and Goldhaber.^{3,4} Its appeal lies in the wide range of electron decay processes encompassed. The electron is assumed to decay in such a way that it leaves a vacancy in the parent atom without interacting with the remaining atomic electrons. The detectable clue left by decay of the initially bound electron would be the subsequent filling of the vacancy in the atomic shell from which it disappeared. For example, emission of characteristic X-radiation and Auger electrons would be readily detectable if the electron vacancy would occur in the K shell of an iodine atom situated in the NaI(Tl) crystal.

Der Mateosian and Goldhaber's experiment consisted of a background spectrum accumulation of 6 1/2 hours with a shielded 4 x 5 in. NaI(Tl) crystal, and the assignment to electron decay of all events within ± 7 kev of an iodine K X-ray calibration peak (28.5 kev.) They calculated a limit on the electron lifetime $\tau_e > 10^{18}$ years.

A portion of this thesis is concerned with an attempt to increase this lower limit on τ_e by reduction of the detector background and by identification and subtraction of background

components due to sources such as K^{40} and Ra^{226} contamination.

Here the search for electron decay is centered at the K-edge energy of 33.2 kev rather than at 28.5 kev, the weighted-average energy of iodine K_{α_1} , K_{α_2} , and K_{β} X-rays formed when the K vacancy is filled by an L or M electron. The reasoning is that the energy which would be released as the iodine atom purged itself of inner shell vacancies would be a total of 33.2 kev, the full K ionization energy. Since the relaxation process occurs in much less time than the 0.25×10^{-6} sec characteristic of NaI(Tl) pulses, the entire characteristic X-ray cascade and/or Auger electrons totaling 33.2 kev would be detected as a single pulse. It can be argued (Appendix I) that the detector response is nearly independent of the particular combination of K, L, M, ..., X-rays and Auger electrons released.

This reasoning must be modified if the decay products of the electron are also captured by the crystal, as would sometimes be the case if one or more of the products were photons. In this situation the 33.2 kev would be added to the coincident photon energy. Accordingly, the escape probability for decay-product photons must be considered in any estimate of the efficiency of a NaI(Tl) crystal for detection of the hole in the iodine K shell.

If in our search for electron decay we look for a particular

set of electron decay products rather than a K shell vacancy alone, some generality is lost. However, we gain the advantage of being able to monitor all electrons in the crystal instead of just the iodine K electrons: there are now 64 candidate electrons per NaI molecule where previously there were 2. Since neutrinos offer negligible chance for detection, we concentrate on the photon as the only detectable electron decay product. Der Mateosian and Goldhaber searched for the process $e^- \rightarrow \nu_e + \gamma$ with the same apparatus used for the K X-ray detection. Because of the greater number of electrons and the higher energy of the photons ($\sim 1/2 mc^2$) it was possible for them to set a more stringent lower limit on the electron lifetime against this decay mode of $\tau_e' > 10^{19}$ years.

On the basis of NaI(Tl) background spectra published by workers concerned with low level counting, it is possible with the der Mateosian and Goldhaber interpretation technique, to infer values of τ_e' up to $\sim 10^{21}$ years.^{5,6,7}

As with the K shell vacancy, this thesis is concerned with the increase of the associated lower limit on τ_e' by further attention to background reduction and the identification of contaminants.

As is evident from the conservation of energy and momentum, if the decay $e^- \rightarrow \nu_e + \gamma$ occurs for a free electron at rest, the

photon and neutrino will have equal and opposite momenta, and will share equally the rest-mass energy of the electron. The resulting photon spectrum will be a delta function at $1/2 mc^2 = 255 \text{ kev}$.

If the decaying electron is bound in an atom, some of the momentum, in general, will be shared by the nucleus. The result is a broadening of the photon spectrum and a downward shifting of its peak from $1/2 mc^2$ to $1/2(mc^2 - E_b)$, where E_b is the binding energy. The more tightly bound the electron, the greater will be the expected broadening of the photon spectrum (Appendix II).

If the process $e^- \rightarrow \nu_e + \gamma$ occurs, in particular for iodine K electrons, 33.2 kev will be released as low energy X-rays and Auger electrons in coincidence with a gamma ray of $\sim 239 \text{ kev}$. The gamma ray, by virtue of its higher energy, has a much greater mean free path in NaI (14 mm) than the radiations associated with the K shell vacancy (0.4 mm). Consequently, the X-rays and Auger electrons are more likely to be trapped in the crystal where they are born, whereas the gamma rays within a centimeter or so of the crystal surface have a good chance of escaping. Hence, we can hope to effect a significant discrimination against background by looking for 33.2 kev events in one crystal in coincidence with 239 kev events in a second, nearby crystal.

Therefore, in addition to aforementioned examination of events at 33.2 kev and 239 kev separately, this thesis includes a study of the two events in coincidence.

II. THE EXPERIMENT

A. Detection of the 33.2 and 255 kev Events Separately

A primary concern in the design of a detection system for electron decay events is to minimize the background. The electron lifetime, τ_e is calculated from the expression $\tau_e = N/(-dN/dt)$, where N is the number of eligible electrons being monitored, and dN/dt is their time rate of change, that is the negative decay rate. Since all unexplained background events near the appropriate energy must be recognized as candidates for electron decay, the rate of these unexplained events is the value assigned to $-dN/dt$. A detector which is sensitive to a large value of τ_e must therefore have a low background ($-dN/dt$) and a relatively large number of electrons (N).

A second consideration in detector design is the efficiency for the radiation in question, a number which depends heavily on assumptions made about the decay process. For example, a detector which would record efficiently the 33.2 kev events associated with the K-shell vacancy would have to permit the escape of decay products which could add their energy to the 33.2 kev. Whether or not the decay products would escape depends on what they are. Neutrinos will escape anything, but photons of the expected energies travel on the average only about 1 cm in NaI before losing energy. Hence a study involving only 33.2

kev events in a large (1 cm) NaI crystal is most sensitive to the cases in which no decay-product photons are involved. As to the detection efficiency for the non-photon decay mode, it is very close to 100% because the low energy X-rays and electrons involved travel less than 0.4 mm in NaI.

The crystal used for these measurements at 33.2 kev was 3 in. long by 3 in. in diameter.

Efficiency and background considerations indicate that the same crystal is suited to 239 kev detection as well. Its efficiency for absorption of this decay radiation is 59% as calculated in Appendix III.

The crystal, mounted in a double end-window housing, was supplied by the Harshaw Chemical Company. A number of low background features were incorporated into the assembly: type 304 stainless steel housing, notably cleaner than the usual aluminum;⁸ MgO reflector, somewhat cleaner than Al_2O_3 ;^{9,10} quartz windows, notably cleaner than glass;¹¹ and a natural potassium content guaranteed by Harshaw to be $<0.8 \pm 0.2$ ppm.

The double window feature permitted a first approximation measurement of the contribution to background made by the notorious radioactivity of the photomultiplier tubes themselves.¹² That is, one tube could be used to measure the background rate with and without the other tube present. However, the difference in rates yields only an approximation to the

background contribution of the tube since scattering and absorption by the tube alter the background from other sources. The double window also permitted coincidence operation of two photomultipliers as a check against tube noise which can be a problem at very low count rates.

The photomultipliers were backed away from the crystal with 3 in. long by 3 in. diameter lucite light pipes. Calculations showed that the consequent reduction in solid angle subtended by the tubes at the crystal, coupled with the attenuation of the radiation by the lucite, reduced the photomultiplier contribution to the background sufficiently to permit the economy of glass tubes as opposed to lower background but more costly quartz tubes.¹³

The photomultipliers selected were 3 1/2 in. E.M.I. type 9531 S. The pins were soldered directly to the voltage divider to avoid the possibility of noisy contacts.

The optically coupled system of phototubes, light pipes, and crystal was sealed in a lucite tube and partially surrounded with triple-distilled mercury (Fig. 1). Aluminized mylar isolated the light pipes and crystal from the mercury. The lucite tube served to contain the components as an integral unit and protect them from contamination within an easily-cleaned surface. All components were carefully washed with detergent and handled with clean rubber gloves during assembly. The need for these

precautions was not experimentally established here, but it has been reported¹⁴ that fingerprints from supposedly clean hands can contribute 20 cpm to background in a large crystal.

With respect to the extensive use of lucite, it should be remarked that no radioactivity has been observed¹⁵ to be associated with lucite except traces from charged radon daughters electrostatically attracted to its surface.

The detector was suspended by strings at the center of a 9 in. thick iron shield with inside dimensions 26 x 26 in. at the base by 27 in. high. The shield was constructed of tiers of iron billets of 3 in. square cross section. It is the same shield used in a different experiment and has been described in detail.¹⁶

Cosmic rays were reduced to $<1/\text{m}^2\text{-min.}$ by virtue of a 2000 ft. underground location in a mine of the Morton Salt Co.

With the two photomultipliers balanced and summed a resolution of 10.7% full width at half maximum (FWHM) was measured for the Cs^{137} gamma when the light pipes were in place. (Some resolution had been sacrificed for the low-noise "S" photocathode of the photomultipliers.)

For accumulation of the medium energy spectrum (near 255 kev) the two photomultipliers were connected in parallel and the output fed through a single preamplifier and amplifier to a 400 channel analyzer. The system was calibrated with a Cs^{137} source

daily except on weekends when the mine was closed. Drift of the system was held to less than 1 per cent during data accumulation.

Since a successful reduction in background by the addition of a 1/8 in. lead lining to an iron shield had been reported in the literature,¹⁷ the iron shield was initially so lined with virgin lead supplied by the National Lead Company under the request that it be of low radioactivity.

In the first arrangement the photomultipliers were coupled directly to the crystal windows, and no mercury was present. Spectra were then accumulated at medium energy (Fig. 2) and high energy (Fig. 3).

In order to check the effectiveness of the lead lining it was removed and a second spectrum accumulated (Fig. 2). A large drop in count rate toward the low end of the medium energy spectrum indicated that the "low-background, virgin lead" contained considerable activity. Since efforts to clean the lead surface were unsuccessful, the lining was not used during the remainder of the experiment.

Insertion of the lucite light pipes brought about a significant background reduction both at medium and high energies (Figs. 2 and 3). When the light pipes were inserted, however, the drop in background was much less than the relative reduction in solid angle subtended at the crystal by the photomultipliers. The implication is that with the light pipes in position the photomultipliers

were no longer a major source of background. For this reason the background change associated with the removal of one photomultiplier was not investigated.

As a final step in reduction of the background, the crystal was surrounded with triple-distilled mercury which occupied the space between the crystal and the lucite envelope and partially covered the light pipes (Fig. 1). The thickness of mercury was .476 cm at the crystal, and .795 cm at the light pipes. Mercury was not extended farther along the light pipes because mercury near the photomultipliers would serve to scatter gamma rays from the photomultipliers toward the crystal. A background spectrum was accumulated at medium energy with the light pipes and mercury in place. As seen in Fig. 4, the reduction in background effected by the mercury was substantial.

This medium-energy background level realized with the light pipes and mercury is the lowest level, per gram of NaI, known by the author to have been achieved in an inert shield. A level lower by a factor of 2 at 255 kev for a 5 x 5 in. NaI crystal in an 18 x 18 in. plastic scintillator anticoincidence guard has been reported by Ellet & Brownell.¹⁸ It is interesting to note that an anticoincidence guard is not necessarily sufficient in itself to insure low background: Perkins, et al¹⁹ reported a background at 255 kev which was higher per gram of NaI than the bottom curve of Fig. 4 by a factor of 3 for a 3 x 3 in. crystal

in a large NaI anticoincidence guard, and by a factor of 4 for a 5 x 5 in. NaI crystal in a large plastic scintillator anticoincidence guard.¹⁹ It is significant for the present experiment that although an anticoincidence guard might be effective with a detector for 255 kev electron-decay photons, it would impair the efficiency of a 33.2 kev K-shell vacancy detector in the sense that it would trap decay-product photons, as discussed in Part I. Therefore efforts were concentrated on reduction of background without an anticoincidence guard to assure that the detector would be effective in the 33.2 kev mode.

The system was operated at medium energy for a total running time of 362 hours (Fig. 5.) A statistical accuracy of $\pm 1.4\%$ ($1/\sqrt{N}$) per channel was achieved.

A high energy spectrum was accumulated for 25.4 hours to help identify sources of background (Fig. 6.) K^{40} is generally present as a source of background. In order to be able to subtract the contribution due to this contaminant it was necessary to measure the crystal response to K^{40} , a determination most readily made with a source containing this isotope (Fig. 6.) 1 kgm of natural potassium was placed near the crystal but outside the mercury shield. The 1.46 mev K^{40} peak in the background is slightly displaced towards lower energies than the peak obtained with the source because in the background spectrum the peak is

riding on a steep slope. As can be seen in Fig. 5 and 6, the K^{40} contributes to the background, not only at the photopeak, but also from the Compton edge all the way to zero energy. The arrangement using an external K^{40} source was chosen because, as will be shown later, most of the K^{40} contributing to the count rate is external to the crystal. A K^{40} spectrum at medium energy was accumulated with the same geometry and was normalized to the high energy K^{40} source spectrum. Thus, when just enough K^{40} radiation was subtracted from the high energy background spectrum to eliminate the 1.46 mev peak, the medium-energy background spectrum was correctly adjusted by subtracting the same fraction of the normalized medium energy K^{40} spectrum. Curve D in Fig. 5 and the lower curve in Fig. 6 show the amount of K^{40} which it is felt from a consideration of the spectral slope can be attributed to this isotope.

The high energy background with K^{40} subtracted appears in Fig. 7. Superimposed on this plot is a spectrum taken with a Ra^{226} source. All the peaks shown in the Ra spectrum are actually due to the Bi^{214} RaC daughter²⁰ which also is a daughter of gaseous 3.8 day Rn^{222} . The appearance of these peaks in the background spectrum indicates the presence of Rn^{222} but not necessarily the Rn^{222} parent, Ra^{226} . The Rn gas could diffuse into the shield from without. Hence, the subtraction of enough of the Ra source spectrum to eliminate the peaks at high energy does not justify the subtraction of an equal amount of the Ra

source spectrum at medium energy where the presence of the 187 kev Ra^{226} line in the background has not been established.

Before subtracting the Ra source spectrum, therefore, we turn our attention to the medium-energy background spectrum (Fig. 5) and look for the 187 kev Ra^{226} peak. It is seen to be present but is nearly lost in the general background. How much of the background is contributed by Ra^{226} as well as its spectral shape is a difficult question. The problem is complicated by the manner in which the mercury is observed to distort the Ra source spectrum below ~ 500 kev (curves B and C, Figl 5) and the uncertainty as to the location of the troublesome Ra^{226} .

The Ra spectrum which was actually subtracted is shown in curve C, Fig. 5. It is the mercury-attenuated Ra test source spectrum scaled to just eliminate the 609 kev Bi^{214} peak. The spectrum subtracted does not contain the characteristic rise at low energies seen in curve B, Figl 5. The medium-energy background spectrum with Ra and K^{40} subtracted (Fig. 8) shows some of the 187 kev Ra^{226} peak remaining, which indicates that the amount of Ra source subtraction was conservative. Judging by the shape of the residual spectrum some of the Ra^{226} gammas do not traverse the mercury. They may arise within the crystal, or more likely, in the photomultiplier tubes. The mercury-attenuated K^{40} source subtraction was conservative for

the same reason: bremsstrahlung from K^{40} radiation could be coming from the photomultipliers and reach the crystal without traversing the mercury.

Care was taken during the subtraction of these Ra and K^{40} backgrounds to avoid the introduction of additional statistical error in the residual background. The source spectra were accumulated with good statistics and smoothed before subtraction.

Two prominent peaks remain in the residual medium-energy background spectrum, one at 425 ± 7 kev, and the other at 503 ± 7 kev (Fig. 8). In the high-energy background spectrum, peaks not associated with the Ra source appear at $1.62 \pm .03$ and $1.91 \pm .06$ mev. Although it would be desirable to identify the sources giving rise to these lines so enabling a further subtraction from background, this has not yet been accomplished despite a preliminary search. For example, among the naturally-occurring radioactive decay series, the thorium series yields photons at 415 kev from $_{82}\text{Pb}^{212}$ and at 511 kev from $_{81}\text{Tl}^{208}$; the uranium series gives a 510 kev photon from $_{86}\text{Rn}^{222}$; and the actinium series gives a 425 kev photon from $_{82}\text{Pb}^{211}$. All of these nuclides can be eliminated as sources of the lines at 425 and 503 kev by the conspicuous absence of strong accompanying lines which would have to appear in the spectrum. The natural decay series offer no photons at 1.62 and 1.91 mev, nor do any

other candidates appear among other naturally occurring nuclides or daughters of nuclides with half-lives $> 10^8$ years. A possible source of the broad 1.91 mev peak is alpha particles of a somewhat higher energy. (The crystal response to alpha particles is not linear.) The peak at 503 kev might be annihilation radiation (511 kev). Contamination by fission products may explain the remaining peaks, but without positive identification their complete spectra remain unknown and cannot be subtracted from background.

A source of background remaining to be discussed is the K^{40} beta spectrum from the $0.8 \pm .2$ ppm natural potassium in the NaI itself. Natural potassium is 0.0119 atom per cent K^{40} . K^{40} emits 31 beta rays per sec. per gram. On the basis of this information and the shape of the K^{40} beta spectrum, it was calculated that a negligible background in the vicinity of 1.3×10^{-4} betas per kev per min. would arise between 0 and 1.35 mev from conservatively assumed 0.6 ppm natural potassium. Compared with 31 beta rays per sec. per gram, K^{40} emits only 3.4 gamma rays per sec. per gram. Gamma rays from internal K^{40} contamination therefore are also negligible. These considerations justify the earlier statement that most of the K^{40} radiation comes from outside the NaI.

The counts making up the residual spectrum (Fig. 8) remain as unexplained events. In accordance with the thesis we choose

to interpret these events as in terms of a limit on the electron lifetime. As was pointed out in Part I, the photon spectrum for bound electrons decaying by $e^- \rightarrow \nu_e + \gamma$ is a delta function at 255 kev for unbound electrons, and is spread out and lowered in energy as the binding energy increased from zero. The most tightly bound are the iodine K electrons, and their spectral broadening is worked out in Appendix II. The great majority of Na and I electrons are much less tightly bound than the iodine K electrons, and their decay photons would likely give a rather sharp spectrum close to 255 kev. In this instance the main cause of broadening is instrumental. The resolution of the system for the 661 kev Cs^{137} gamma is 10.7%. An estimate of the resolution at 255 kev can be made on the assumption that resolution is determined by photon statistics, namely

$$\begin{aligned} (\text{Resolution at 255 kev}) &= \sqrt{\frac{661}{255}} \times (\text{Resolution at 661 kev}) \\ &= 17.2\% \text{ FWHM.} \end{aligned}$$

The portion of the residual background near 255 kev after Ra and K^{40} were subtracted was plotted together with the approximate theoretical electron decay photon spectrum (Fig. 9). The 17.2% resolution gives a 44 kev width to the theoretical peak.

If, as an illustration, all the events of Fig. 9 in the 44 kev interval centered at 255 kev are assumed to be electron-decay photons, τ_e can be calculated as 3×10^{20} year. (Appendix IV)

This value of τ_e is low for several reasons, among which is the fact that the calculation was done under the assumption that the only source of decaying electrons is the NaI. Electrons in the surrounding mercury and other materials are equally likely to decay and bathe the crystal in an external flux of 255 kev photons as well.

The value $\tau_e > 3 \times 10^{20}$ year is much too low for the following additional reason. The residual count rate in the 44 kev band at 255 kev cannot possibly be due exclusively to decaying electrons. A portion of the rate in this band is made up of the compounded Compton tails belonging to photopeaks at higher energy, particularly at 425 and 503 kev. In addition, the nearly featureless structure of the residual spectrum at 255 kev indicates a smoothly rising background of bremsstrahlung, backscatter, and indistinct components against which a substantial 255 kev photopeak would be very conspicuous. The slightly concave character of the residual spectrum further emphasizes the lack of such a peak. Statistical fluctuations ($\pm 1.4\%$) in the counts per channel across the 44 kev band are not enough to conceal more than a few per cent of the electron 255 kev decay photon peak.

The number of counts which can be attributed to electron decay is not clearly specified by the residual spectrum (Fig. 9). However, in the absence of any clearly defined structure it seems

reasonable to associate such a limit with statistical fluctuations which could obscure the spectrum B of Fig. 9 which is expected from electron decay as described earlier. We, therefore, take the fluctuation in the number of counts in the energy range 44 kev surrounding the 255 kev peak. This number ($= 3.7 \times 10^3/\text{yr}$) is somewhat conservative because the background is not constant but rises with decreasing energy. On this basis the lifetime limit is $\tau_e' > 5 \times 10^{22}$ years.

The same detector, with mercury and light pipes in place, was used to accumulate a low-energy spectrum (near 33.2 kev). Because of the greater danger of interference from photomultiplier tube noise at low energy, separate preamplifiers and amplifiers were used for the two tubes and the pulse-height analyzer was gated with a coincidence pulse between the two tubes. An upper-level discriminator rejected very large pulses which threatened by means of delay-line reflections to introduce counts at low energy. Visual observation of the two tube outputs displayed simultaneously on a dual-beam oscilloscope confirmed that the system was functioning properly in the coincidence mode.

The frequency of calibration and the system stability were the same as at medium energy. The calibration was performed with the 74 kev Pb X-ray from the Ra source. The mercury had to be partially removed for each calibration because it so

greatly attenuated the 74 kev X-ray.

The resolution for the 74 kev X-ray was measured as 35% (Fig. 10). With the same technique used to estimate the resolution at 255 kev, the resolution at 33.2 kev was estimated at $\sqrt{74/33.2} \times 35\% = 52\%$. The full width at half maximum of the expected 33.2 kev peak is $0.52 \times 33.2 \text{ kev} = 17 \text{ kev}$ (Fig. 11).

A spectrum was accumulated at low energy for a total run time of 110 hours to give better than 7.6% statistics in each channel (Fig. 11). No subtraction of identified background components has been made in the spectrum of Fig. 11. The behavior of the Ra and K source spectra was not investigated at low energy. However, the Compton tail associated with the 1.46 mev K^{40} photopeak can be subtracted because it arises from 1.46 mev photons which penetrate the mercury and lose only a little energy in the NaI. From Fig. 5 it can be seen that this tail is nearly flat. (The Compton tail for 1.46 mev photons continues to rise slowly as the energy decreases.²¹) The bremsstrahlung is attenuated by the mercury. The Compton tail compounded from the many photopeaks of the Ra source is lost under the peaks between 74 and 400 kev (Fig. 5 curve C) but is at least as high at 33.2 kev as it is near 500 kev, below the 609 kev Bi^{214} photopeak. From curve C, Fig. 5, this level is 0.0052 counts per kev per min. The important point in this discussion is that the various known backgrounds do not exhibit

structure of the kind which would tend to obscure a possible electron decay signal. As a conservative estimate of the electron lifetime from these data, we ascribe to electron decay the statistical fluctuation in the number of counts seen in the 17 kev energy region around the 33.2 kev K-peak.

This is conservative because the experimental curve is concave upwards whereas the decay curve is predicted to be concave downward. The number of events ascribable in this limit calculation to electron decay is 2300/year corresponding to a value of $\tau_e > 4 \times 10^{21}$ years.

B. Detection of the 33.2 and 239 kev Events in Coincidence

The arrangement for detecting the coincidence of the 33.2 kev radiation (subsequent to the filling of an iodine K-shell vacancy) and the $\sim$ 239 kev electron-decay photon consisted of two 3 in. thick x 11 1/2 in. diameter Harshaw NaI(Tl) crystals mounted in close proximity (Fig. 12). The crystals were housed in 0.019 in. thick stainless steel with 0.106 in. thick MgO reflector. Seven three inch photomultipliers viewed each crystal. The photomultiplier leads were soldered directly to the voltage dividers so eliminating the normally radioactive tube bases. Because of a slight bow in the crystal faces their separation varied from 1/16 in. at the center to 5/16 in. at the periphery.

With the photomultipliers balanced, the resolution of lower crystal (#1) was measured at 8.6% FWHM for Cs¹³⁷ with the source scanning over the "flat" face during spectrum accumulation. The resolution of the upper crystal (#2) was measured as 8.7% by the same technique. The crystals were situated in the same iron shield used previously for the 3 x 3 in. crystal.

The output of each crystal was fed through its own pre-amplifier and amplifier. The direct output of each amplifier was connected through a delay line to a dual-beam oscilloscope, one output to each beam (Fig. 13). The differential discriminator on amplifier #1 gave a triggered pulse for events between 27.2

and 39.2 kev, while that for #2 responded to events between 190 and 450 kev. The triggered pulses were fed through a coincidence circuit, and the coincidence output triggered a scaler, the scope, and a scope camera. A pulser check showed that the system functioned properly for all combinations of pulse heights within the discriminator windows.

The system was calibrated with a Ra source placed between the crystals on the common axis. The 74 kev Pb X-ray was used for crystal #1, the 242 kev Bi²¹⁴ gamma was used for crystal #2. During the early runs the system was calibrated three times a week, but it became evident that it was sufficiently stable (<3% drift) to permit a calibration interval of one week.

The background spectrum at low energy in crystal #1 is shown in Fig. 14 with the 12 kev window. The shape of the 33.2 kev peak, also shown, was estimated in the same manner as for the 3 x 3 in. crystal.

The background spectrum at medium energy in crystal #2 is shown in Fig. 15 with the 260 kev window. A Ra calibration is superimposed.

The rate of pulses in the 27.2 to 39.2 window was ~ 19 per min. In the 190 to 450 kev window the rate was ~ 450 per min. The coincidence rate was ~ 40 per hr. Fluctuations in these rates of $\pm 5\%$ were common. (The accidental coincidence

rate is calculated as $\sim 8 \times 10^{-3}$ per hr.)

The heights of the two pulses on each frame taken by the camera were measured. The pulse height (X) from the medium-energy crystal was recorded beside the pulse-height (Y) from the low energy crystal. Each coincident event, then, corresponded to a point in X, Y space. This space was divided up into "bins" and a computer program was written to sort the pairs into the appropriate bins. The bin size was chosen for a favorable balance between resolution and statistics. The bins were made smaller around the periphery defined by the window boundaries, because the differential discriminators did not cut off sharply at the limits, and it was desirable to see how deeply these fuzzy edges invaded the enclosed two-dimensional space.

The pairs were fed to the computer as a continuous string of numbers on punch cards.* The print-out was in the form of a matrix, the elements of which corresponded in their arrangement to the rows and columns of bins. Along the top and down the left side, between the columns or rows, were written the bin boundaries in kev. Along the bottom were printed the column totals, and down the right side, the row

*To insure against a mismatching of pairs, should half of a pair be dropped somewhere, the pairs were punched 12 to a card, always beginning a card with the Y member of a pair. The computer rejected cards with other than exactly 24 numbers (12 pairs) and indicated the rejection.

totals (Table I.)

The density distribution of Table I is not the two-dimensional-energy-space spectrum expected from the decay of iodine K electrons by the mode $e^- \rightarrow \nu_e + \gamma$. The expected spectrum would be a band of high relative density running from left to right and centered vertically. This concentration would reflect the 33.2 kev peak. The density in the band would be greatest at 239 kev on the horizontal scale and would fall off symmetrically on both sides in the horizontal direction according to the distribution given in Appendix II for the 239 kev peak.

It is a matter of some interest to understand the source of the counts in Table I since this knowledge could be helpful in background reduction. Among the contributors could be multiple Compton processes. Some are shown in Fig. 16. Since a 33.2 kev photon does not have an appreciable chance ($\sim 10^{-3}$) of penetrating the two thicknesses each of stainless steel and MgO, the more likely processes are those in which the energy is carried between crystals by more energetic photons (Fig. 16). A Compton electron could be the inter-crystal carrier if it had sufficient energy to penetrate at least one thickness of stainless steel and MgO. (Because of the character of the Compton

process the photon giving the energy to this electron would have to be greater than 1.6 mev to give the electron the necessary 1.4 mev needed for the penetration.)

Not much can be done about photons crossing the gap without interfering with possible electron-decay photons, but one could discriminate against electrons with a piece of lucite inserted between the crystals. One eight inch of lucite is only 0.04 mean-free-paths for 239 kev photons, whereas electrons going straight through would lose 800 kev. When 1/8 in. of lucite was inserted between the crystals, there was no statistically significant change in the coincidence rate.

If photons at ~ 239 kev are carrying energy across the gap to cause the coincidence events, insertion of 1/8 in. of lead between the crystals should reduce the coincidence rate to $\sim 10\%$ of its value. When 1/8 in. of lead was inserted, the coincidence rate fell to 33% of its former rate. (The windows for the lead and lucite tests were ± 5 kev in crystal #1 and ± 30 kev in crystal #2.) The insertion of 1/2 in. of lead in the gap would attenuate 239 kev photons to $\sim 0.02\%$ of their intensity, but 1/2 in. of lead actually reduced the open gap coincidence rate to 12% of its initial value. These

tests indicate the participation of energetic gamma rays in the background coincidence rate. The energetic photons capable of feeding the coincidence counter were actually present (Fig. 17).

When crystal #2 was removed and placed on top of the shield, no coincidence counts were registered in 15.5 hours. This test demonstrated that the coincidence counts were not being generated by cross-talk in the electronics.

A portion of Table I has been included in a rectangular box. The horizontal boundaries of this box define the full width at half maximum of the theoretical 33.2 kev peak. The vertical boundaries could not be drawn to define the full width at half maximum of the theoretical 239 kev peak because of the invasion of the lower discriminator into the spectrum above 190 kev. (This invasion is apparent in the first three columns of the table.) The vertical boundaries as drawn define 60% of the full width at half maximum of the 239 kev peak. Since the distribution expected for electron decay was not evident in the data displayed in the table it was considered reasonable to estimate the lower limit for electron stability by the use of statistical fluctuations as before. A total of 510 counts were registered in the box in 76.7 hours. The lifetime limit τ_e was calculated as in Appendix IV with the following differences: for $-dN/dt$, $\sqrt{510} / (76.7\text{hr} \times .60)$ was

used, 18.8 kg was used for M, and the efficiency applied was $\epsilon_L = 12\%$ (Appendix III). On this basis $\tau_e'' > 4 \times 10^{21}$ year. This number will be improved considerably by analysis of more of the available data.

It is interesting to note the approximate correspondence between the column totals of Table I and the medium-energy background spectrum for crystal #2 (Fig. 15). The correspondence breaks down below 210 kev, however, where the column totals begin to rise very rapidly as energy decreases. (It is to be kept in mind that the first three columns represent undersized bins.) This rise below 210 kev is about where backscatter from crystal #1 would be expected to contribute heavily.

TABLE I

Distribution of Coincidences in Two-dimensional
Energy Space

Kev	180	190	200	210	230	250	270	290	310	330	350	370	390	410	430	450	460	470	480
25.7	0	12	32	23	28	21	26	34	37	24	16	18	17	13	26	10	9	1	347
27.2	1	18	25	31	31	15	26	22	35	19	11	20	17	31	22	11	7	0	342
28.7	0	36	41	58	50	48	62	59	62	46	41	37	36	36	48	10	21	1	692
31.7	3	24	58	73	46	64	48	55	67	50	34	31	32	28	42	18	6	1	680
34.7	4	32	64	51	49	71	69	65	54	49	29	35	27	32	42	9	11	2	695
37.7	1	14	22	23	18	26	23	21	32	13	9	13	16	10	19	4	5	0	269
39.2	1	0	2	2	4	1	2	4	7	3	6	5	0	1	3	0	0	0	41
40.7	10	136	244	261	226	246	256	260	294	204	146	159	145	151	202	62	59	5	

III. SUGGESTED ADDITIONAL EXPERIMENTATION

Toward the end of the coincidence run with crystals #1 and #2 a drop of 25% occurred in the coincidence rate and persisted for one week after which it was necessary to terminate the experiment. The singles rates in the crystals were relatively unchanged, and the electronics checked out normally. During this week of low count rate the fan used to pull air down the mine shaft had been shut down more than usual. The thought occurs that the level of 3.8 day Rn^{222} in the mine might have fallen off enough to account for the drop in count rate.

That the singles rates were relatively unaffected is not inconsistent with the change in coincidence rate since a large fraction of the singles rate is caused by radiation near the photomultipliers. The portions of the crystals near the surfaces facing the gap are shielded by NaI from surrounding radiation. A 25% change in the small fraction of radiation entering from the gap would not affect the singles rate appreciably, but could easily cause a 25% change in the coincidence rate which is strongly dependent on radiation in the gap. Radon gas in the gap could be a major cause of the coincident events recorded. To investigate this possibility it might be worthwhile to isolate the shield with an air-tight bag and monitor the coincidence rate for a few Rn^{226} half lives.

After the crystals #1 and #2 are isolated from radon, a further background reduction could undoubtedly be effected through the techniques successful in reducing background in the 3 x 3 in. crystal, namely the use of light pipes and mercury.

With respect to the 3 x 3 in. crystal, there is reason to believe that more mercury (up to an inch thick) and longer light pipes would lower the background significantly.* It might be advisable to abandon the low-noise "S" photocathode on the photomultiplier tubes in favor of a higher photocathode sensitivity. The advantages of the resulting improvement in resolution would be two-fold. First, narrower instrumental peaks would reduce the necessary window width and associated count rate. Second, identification of background contributions would be simplified by the smaller uncertainty in the photopeak energies.

*As operated, the background due to radioactivity of the photomultiplier tubes was not limiting. An increased mercury shield would change this situation and require the increased isolation provided by longer light pipes.

APPENDIX I

The Response of NaI(Tl) to a Vacancy in the Iodine K Shell

The following argument can be made for the lack of strong dependence of NaI(Tl) response on the particular combination of K,L,M... X-rays and Auger electrons subsequent to the appearance of a vacancy in the K shell of iodine.

It has been shown that NaI(Tl) responds linearly for X-rays ranging from 411 kev down to as low as 2 kev.²² The K,L, and M absorption edges for iodine fall roughly at 33, 5, and 1 kev respectively. When the filling of the K shell produces an X-ray its energy must be $33 - 5 = 28$ kev for K_{α} , $33 - 1 = 32$ kev for K_{β} , etc. When the filling of the L shell results in X-rays their energies must be at least $5 - 1 = 4$ kev. After the L shell is filled only 1 kev remains to be lost. Clearly for the case of characteristic X-ray emission most of the de-excitation energy ($32/33$) is released in quanta above 2 kev where the NaI(Tl) response is known to be linear.

The K fluorescence yield for iodine is 88%,²³ which means that Auger electrons replace K X-rays 12% of the time. Auger electrons resulting from a vacancy in the K shell can be designated as K-XY, where X is the shell supplying the electron to the hole in the K shell, and Y is the shell from which the Auger electron is ejected. In iodine the K-LL electron has an energy of $33 - 5 - 5 = 23$ kev; the K-LM energy is

$33 - 5 - 1 = 27$ kev, etc. The corresponding K-MY energies will be still higher. L shell vacancies can be filled by L X-rays (at least 4 kev) or conceivably L-XY Auger electrons, the lowest energy of which would be the L-MM at $5 - 1 - 1 = 3$ kev. The M-XY electrons would have less than 2 kev energy. The worst imaginable sequence, i.e. the sequence yielding the least energy in quanta of more than 2 kev, would be: K-LL (23 kev), L-MM (3 kev), L-MM (3 kev)...., for a total of 29 kev within the known linear region of NaI(Tl).

In short, a vacancy in the K shell of iodine will be recorded by a NaI(Tl) crystal as an average energy release of close to 33 kev.

APPENDIX II, courtesy of Professor L. L. Foldy

Spectrum for Decay of a Bound Electron

We consider a process in which a free electron decays into a photon and neutrino:

$$e^- \rightarrow \gamma + \nu$$

If such a process occurs for an electron at rest, clearly the γ and ν have equal and opposite momenta and the energy of each is half the electron rest energy. The γ -spectrum then has a δ -function peak at $1/2 mc^2$

If the electron is bound in the K shell of an atom with binding energy E_b , the photon spectrum will now be continuous. We wish to estimate the effect of this binding on the photon spectrum. Since we do not know how to write a proper interaction for this decay process, we cannot obtain an unambiguous result. At best we can specify certain of the factors which will in general be present in the transition probability. Denoting the photon momentum by $\hbar k$ and the neutrino momentum by $\hbar v$, conservation of energy requires:

$$mc^2 - E_b = \hbar v c + \hbar k c$$

The phase space factor for the decay will be proportional to:

$$v^2 d\Omega_v k^2 d\Omega_k dk$$

In addition we expect the square of a transition matrix element

M. One factor occurring in M will be

$$\int \psi_e(\vec{r}) e^{-i(\vec{\nu} + \vec{k}) \cdot \vec{r}} d\vec{r} = \phi(\vec{\nu} + \vec{k})$$

whose square is the momentum probability distribution of the electron in the bound atomic state.

In addition the creation of the photon involves a factor in M of the form $\frac{1}{\sqrt{k}}$. There may be additional factors which depend on the form of the "current" which interacts with the electromagnetic field. Such factors cannot be ascertained without a detailed theory of the interaction. They will not generally vary rapidly with photon energy. It is convenient to assume that they simply cancel the $\frac{1}{\sqrt{k}}$ from photon creation so that the resultant photon energy distribution is symmetric about the midpoint of the spectrum.

Under these assumptions the photon energy distribution is of the form

$$\int |\phi(\vec{\nu} + \vec{k})|^2 d\Omega_k \nu^2 k^2 dk$$

We therefore need the integral

$$\int |\phi(\vec{\nu} + \vec{k})|^2 d\Omega_k$$

The K shell electron wave function is of the form

$$\psi_e(r) = e^{-\alpha r}$$

where

$$\alpha = \frac{Z}{a_0}$$

Then

$$\phi(\vec{r} + \vec{k}) \propto \frac{1}{[\alpha^2 + (\nu + k)^2]^2}$$

and

$$\begin{aligned} \int |\phi|^2 d\Omega_\nu &= \int_0^\pi \frac{\sin \theta d\theta}{[\alpha^2 + \nu^2 + k^2 + 2\nu k \cos \theta]^4} \\ &= \frac{1}{2\nu k} \left\{ \frac{1}{[\beta - 2\nu k]^3} - \frac{1}{[\beta + 2\nu k]^3} \right\} \\ &= \frac{6\beta^2 + 8\nu^2 k^2}{[\beta^2 - (2\nu k)^2]^3} \end{aligned}$$

where $\beta = \alpha^2 + \nu^2 + k^2$.

Now

$$\begin{aligned} \beta^2 - (2\nu k)^2 &= [\alpha^2 + \nu^2 + k^2 + 2\nu k - 2\nu k]^2 - 4\nu^2 k^2 \\ &= [\alpha^2 + (\nu + k)^2]^2 - 4\nu k [\alpha^2 + (\nu + k)^2] \\ &= [\alpha^2 + (\nu + k)^2][\alpha^2 + (\nu - k)^2] \end{aligned}$$

Now from energy conservation

$$\nu + k = \frac{mc^2 - E_b}{hc} = \epsilon$$

a constant, so

$$\beta^2 - (2\nu k)^2 = [\alpha^2 + \epsilon^2][\alpha^2 + (\nu - k)^2]$$

Also

$$\begin{aligned} 3\beta^2 + 4\nu^2 k^2 &= 3[\alpha^2 + (\nu + k)^2][\alpha^2 + (\nu - k)^2] + 8\nu^2 k^2 \\ &= 3[\alpha^2 + \epsilon^2][\alpha^2 + (\nu - k)^2] + 8\nu^2 k^2 \end{aligned}$$

Thus the energy distribution of photons will be proportional to

$$\frac{3[\alpha^2 + \epsilon^2][\alpha^2 + (\nu - k)^2] + 8\nu^2 k^2}{[\alpha^2 + (\nu - k)^2]^3} \cdot \nu k^2 dk$$

or

$$\frac{3[\alpha^2 + \epsilon^2][\alpha^2 + (\epsilon - 2k)^2] + 8(\epsilon - k)^2 k^2}{[\alpha^2 + (\epsilon - 2k)^2]^3} (\epsilon - k)^2 k^2 dk$$

One may use this formula with all energies measured in units of mc^2 . In this case

$$\alpha = \frac{Z}{137}$$

$$\epsilon = 1 - \frac{E_b}{mc^2} \simeq 1 - \frac{1}{2} \left(\frac{Z}{137} \right)^2$$

The distribution functions is symmetrical about $\epsilon/2$ (Fig. 17).

APPENDIX III

Estimated Efficiencies of the Detectors

A. The 3 x 3 inch crystal.

The efficiency (ϵ_s) of the small crystal for 239 kev photons born uniformly throughout the crystal volume can be written as $\epsilon_s = 1 - \bar{P}_s$, where $\bar{P}_s$ is the average probability that a photon will escape the small crystal without losing its total energy. For photons of 239 kev, we assume that a single interaction suffered by the photon on its way to surface leads to total energy loss within the crystal. The assumption is completely valid for $\sim$ half of the photons inasmuch as $\sim$ half of the total attenuation cross section at 239 kev is photoelectric.²⁴ The remaining half of the photons, after having lost energy in a single Compton interaction, fall prey to a rapidly rising photoelectric cross section. Since there will be some photons escaping after one or more Compton interactions, however, the approximation leads to a slightly higher value of ϵ_s than is actually the case.

Under this approximation, we can write

$$\bar{P}_s = \frac{\int_V \int_{\Omega} e^{-\mu|\vec{r}|} d\Omega dV}{\int_V \int_{\Omega} d\Omega dV}$$

where μ is the total linear attenuation coefficient for 239 keV photons in NaI, $|\vec{s}|$ is the path length to a point on the crystal surface from a volume element within, and $d\Omega$ and dV are solid angle and volume elements respectively.

The integral can be split into two sections, one taking in the curved surface of the cylinder and one the plane surfaces. If we consider the curved surface first and use cylindrical coordinates (r, θ, z) with z measured from the midpoint along the cylinder axis, we can describe a point within the volume by (r, θ, z) and a point on the curved surface by (R, θ', z') where R is the radius of the cylinder and θ' and z' are the surface coordinates and have the same origin as θ and z . The volume element becomes $dV = r dr d\theta dz$, and the solid angle element becomes

$$d\Omega = \frac{R d\theta' dz'}{|\vec{s}|^2} \cos(\vec{R}, \vec{s})$$

where $R d\theta' dz'$ is an element of surface area, and $(\vec{R}, \vec{s})$ is the angle between $\vec{R}$ and $\vec{s}$.

It can be shown by simple geometrical considerations that

$$|\vec{s}| = [(z' - z)^2 + r^2 + R^2 - 2rR \cos(\theta - \theta')]^{1/2}$$

and that

$$\cos(\vec{R}, \vec{s}) = \frac{R^2 + |\vec{s}|^2 - r^2 - (z' - z)^2}{2 R |\vec{s}|}$$

Also it is clear that

$$\int_V \int_{\Omega} d\Omega dV = 4\pi(\pi R^2 h) = 4\pi^2 R^2 h$$

where h is the height of the cylinder. Therefore

$$\bar{P}_{sc} = \frac{1}{4\pi^2 R h} \int_{z=-\frac{h}{2}}^{+\frac{h}{2}} \int_{r=0}^R \int_{\theta=0}^{2\pi} \int_{z'=-\frac{h}{2}}^{+\frac{h}{2}} \int_{\theta'=0}^{2\pi} \frac{[R - r \cos(\theta - \theta')] r e^{-\mu|\vec{s}|}}{|\vec{s}|^3} d\theta' dz' d\theta dr dz$$

where $\bar{P}_{sc}$ is the escape probability from the curved surface.

We can eliminate the integration over θ by restricting the volume integration to a wedge of angle $d\theta$ and length $\frac{h}{2}$.

Let θ for this wedge be $\theta = 0$. In addition, the symmetry is such that θ' need go only to π , provided the integral is multiplied by 2.

Then we have

$$\bar{P}_{sc} = \frac{2}{\pi R h} \int_{z=0}^{+\frac{h}{2}} \int_{r=0}^R \int_{z'=-\frac{h}{2}}^{+\frac{h}{2}} \int_{\theta'=0}^{\pi} \frac{(R - r \cos \theta') r e^{-\mu|\vec{s}|}}{|\vec{s}|^3} d\theta' dz' dr dz$$

This integral was programed for numerical evaluation by the UNIVAC 1107. The trapezoidal rule was used with very small increment spacing in regions of rapid variation of the integrand, and larger spacing elsewhere. The program appears

in Fig. 19. The resulting $\bar{P}_{sc}$ is 30%.

Similar reasoning leads to the following expression for the escape probability, $\bar{P}_{spn}$, from the near plane surface:

$$\bar{P}_{spn} = \frac{2}{\pi R^2 h} \int_{z=0}^{\frac{1}{2}} \int_{r=0}^R \int_{\theta=0}^{\pi} \int_{r'=0}^R \frac{r z r' e^{-\mu |\vec{s}'|}}{|\vec{s}'|^3} dr' d\theta' dr dz$$

where $|\vec{s}'| = (Z^2 + r^2 + r'^2 - 2rr' \cos\theta')^{1/2}$ and Z is now measured from the near plane surface rather than from the midpoint of the crystal. The same expression yields the escape probability, $\bar{P}_{spf}$, for the far surface if z is replaced by (h-z). The programs appear in Figs. 20 and 21. The results are $\bar{P}_{spn} = 11\%$, and $\bar{P}_{spf} = 0.3\%$. In summary,

$$\bar{P}_s = \bar{P}_{sc} + \bar{P}_{spn} + \bar{P}_{spf} = 41\%$$

$$\text{and } \epsilon_s = 1 - \bar{P}_s = 59\%$$

A crude estimate shows this result to be more or less as expected.

B. The 3 x 11 1/2 inch crystal

Events of interest in the case of the two crystals of Fig. 12 are those in which a 239 kev photon born in the lower crystal escapes that crystal with no interaction and enters the upper crystal where it is captured. In essence, the problem

is to find the average probability $\bar{P}_{lpn}$ that a 239 kev photon born somewhere within the volume of the $3 \times 10 \frac{1}{2}$ inch crystal will escape the upper surface without interaction.

From Fig. 12 and a knowledge of the relevant gamma mean free path (1.36 cm) a capture efficiency of 90% in the upper crystal seems reasonable. The efficiency ϵ_L can therefore be written, as $\epsilon_L = 0.9 \bar{P}_{lpn}$ where $\bar{P}_{lpn}$ is the probability integral ($\bar{P}_{spn}$) calculated previously for the small crystal, but with appropriate changes in the size parameters. The program appears in Fig. 22 and gives the result $\bar{P}_{lpn} = 13\%$, from which $\epsilon_L = 0.9 \times 13\% = 12\%$.

APPENDIX IV

Sample Calculation of τ_e'

$$\tau_e' = \frac{N}{-(dN/dt)} \epsilon_s$$

where N is the number of electrons in the crystal, $(-dN/dt)$ is the count rate in 44 kev at 255 kev, and ϵ_s is the detection efficiency of the 3 x 3 in. crystal for internally born 255 kev photons (Appendix III).

$$N = A \frac{M}{m} n$$

where A is Avogadro's number, M is the mass in grams of the NaI, m is the molecular weight of NaI (149.92), and n is the number of electrons per NaI molecule (64).

$$M = \rho V = 3.67 \frac{\text{g}}{\text{cm}^3} \times \pi \times (3.81)^2 \times 7.62 \text{ cm}^3$$

$$M = 1278 \text{ g}$$

$$N = \frac{6.03 \times 10^{23} \times 1278 \times 64}{149.92} = 3.28 \times 10^{26}$$

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$$-\frac{dN}{dt} = \frac{0.026}{\text{kev min}} \times 44 \text{ kev} \times 5.25 \times 10^5 \frac{\text{min}}{\text{yr}}$$
$$= 6.00 \times 10^5 \text{ yr}^{-1}$$

$$\epsilon_s = 0.59$$

$$\tau_e' = \frac{3.28 \times 10^{26}}{6.00 \times 10^5} \times 0.59 = 3.23 \times 10^{20} \text{ yr}$$

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Fig. 1 -- The 3 x 3 in. crystal detector

Fig. 5 3 x 3 IN. CRYSTAL

MED. ENERGY

A: BACKGROUND

B: Ra CALIBRATION

WITHOUT Hg

C: Ra²²⁶ SOURCE

WITH Hg

D: K⁴⁰ SOURCE

WITH Hg

Fig. 9
3 x 3 IN. CRYSTAL

A: BACKGROUND NEAR 255 KEV

B: THEORETICAL ELECTRON-DECAY GAMMA SPECTRUM

Fig. 10 3 x 3 IN. CRYSTAL

RESOLUTION FOR
THE 74 KEV Pb X-RAY

Fig. 11
3 x 3 IN. CRYSTAL
LOW ENERGY

Fig. 12 -- The coincidence detector

Fig. 13

Fig. 15 3 x 11 1/2 IN. CRYSTAL #2
MED. ENERGY

Fig. 16 Source of coincidence background

ALG EFNCY $\bar{P}_{sc}$
ALGOL

MARCH 21, 1965

INTERFACE FEBRUARY 15, 1965

PASS2

```

1 REAL DR, R, W, V, DZ, Z, Q, T, DZP, ZP, DTP, S, F, P, TP, U, PRAR$
  LEVEL 1
2 INTEGER N, M, L, K$
3 N = 1$ DR = .3310$ R = 3.6445$ W = 0$
4 LOOP5.. M = 1$ V = 0$ nZ = .3310$ Z = 3.6445$
5 LOOP4.. Q = 0$ T = Z$ DZP = .2$ ZP = Z + .1$
6 LOOP3.. L = 1$ LOOP2.. K = 1$ DTP = .0995$
7 S = SQRT((ZP - Z)*(ZP - Z) + 14.5 + R*(R - 7.62))$
8 F = (3.81 - R)*R*(EXP(-.7345))/(S*S*S)$
9 P = .0645F$ LOOP1.. K = 2K + 1$ TP = .0332K$
10 S = SQRT((ZP - Z)*(ZP - Z) + 14.5 + R*(R - 7.62COS(TP)))$
11 F = (3.81 - R*COS(TP))*R*(EXP(-.7345))/(S*S*S)$
12 P = P + F*DTP$ DTP = 2DTP$ IF TP LSS 2 THEN GOTO LOOP1$
13 IF ZP LSS Z THEN GOTO X1$ L = 2L + 1$ ZP = Z + .1L$
14 T = T + DZP$ IF T GTR 3.81 THEN GOTO X20$ Q = Q + P*DZP$
15 IF DZP GTR .25 THEN GOTO X21$ DZP = .3$ GOTO LOOP2$
16 X21.. DZP = 2DZP$ GOTO LOOP2$ X20.. DZP = DZP - T + 3.81$
17 Q = Q + P*DZP$ T = 0$ DZP = .2$ ZP = Z - .1$
18 GOTO LOOP3$ X1.. L = 2L + 1$ ZP = Z - .1L$ T = T + DZP$
19 U = Z + 3.81$ IF T GTR U THEN GOTO X2$ Q = Q + P*DZP$
20 IF DZP GTR .25 THEN GOTO X3$ DZP = .3$ GOTO LOOP2$
21 X3.. nZP = 2DZP$ GOTO LOOP2$ X2.. DZP = nZP - T + U$
22 Q = Q + P*DZP$ V = V + Q*nZ$ IF M GTR 8 THEN GOTO X5$
23 M = 2M + 1$ Z = 3.81 - .1655V$ IF DZ GTR .4 THEN GOTO X4$
24 nZ = .4965$ GOTO LOOP4$ X4.. DZ = 2nZ$ GOTO LOOP4$
25 X5.. W = W + V*DR$ IF N GTR 8 THEN GOTO X6$ N = 2N + 1$
26 R = 3.81 - .1655N$ IF DR GTR .4 THEN GOTO X7$ DR = .4965$
27 GOTO LOOP5$ X7.. DR = 2DR$ GOTO LOOP5$
28 X6.. PRAR = W/45.5$ WRITE('PRAR', PRAR)$
29 FINISH

```

PRAR

3.0146,-01

Fig. 19 -- Program for $\bar{P}_{sc}$.

ALG EFFCY $\bar{P}_{SPN}$
AI GOL

MARCH 21, 1965

INTERFACE FEBRUARY 15, 1965

PASS2

```

1  REAL DR, R, W, V, DZ, Z, Q, T, DRP, RP, DTP, S, F, P, TP, PBAR$
   LEVEL 1
2  INTEGER N, M, L, K$
3  N = 1$ DR = .3310$ R = 3.6445$ W = 0$
4  LOOP5.. M = 1$ V = 0$ DZ = .3310$ Z = .1655$
5  LOOP4.. Q = 0$ T = R$ DRP = .2 $ RP = R + .1$
6  LOOP3.. L = 1$ LOOP2.. K = 1$ DTP = .0995$
7  S = SQRT(Z*Z + R*R + RP*RP - 2*R*RP)$
8  F = R*Z*RP*(EXP(-.734*S))/(S*S*S)$
9  P = .0645F$ LOOP1.. K = 2K + 1$ TP = .0332K$
10 S = SQRT(Z*Z + R*R + RP*RP - 2*R*RP*COS(TP))$
11 F = R*Z*RP*(EXP(-.734*S))/(S*S*S)$
12 P = P + F*DTP$ DTP = 2DTP$ IF TP LSS 2 THEN GOTO LOOP1$
13 IF RP LSS R THEN GOTO X1$ L = 2*L + 1$ RP = R + .1L$
14 T = T + DRP$ IF T GTR 3.81 THEN GOTO X20$ Q = Q + P*DRP$
15 IF DRP GTR .25 THEN GOTO X21$ DRP = .3$ GOTO LOOP2$
16 X21.. DRP = 2*DRP$ GOTO LOOP2$ X20.. DRP = DRP - T + 3.81$
17 Q = Q + P*DRP$ T = 0$ DRP = .2$ RP = R - .1$
18 GOTO LOOP3$ X1.. L = 2*L + 1$ RP = R - .1L$ T = T + DRP$
19 IF T GTR R THEN GOTO X2$ Q = Q + P*DRP$
20 IF DRP GTR .25 THEN GOTO X3$ DRP = .3$ GOTO LOOP2$
21 X3.. DRP = 2*DRP$ GOTO LOOP2$ X2.. DRP = DRP - T + R$
22 Q = Q + P*DRP$ V = V + Q*DZ$ IF M GTR 8 THEN GOTO X5$
23 M = 2*M + 1$ Z = .1655*M$ IF DZ GTR .4 THEN GOTO X4$
24 DZ = .4965$ GOTO LOOP4$ X4.. DZ = 2DZ$ GOTO LOOP4$
25 X5.. W = W + V*DR$ IF N GTR 8 THEN GOTO X6$ N = 2N + 1$
26 R = 3.81 - .1655N$ IF DR GTR .4 THEN GOTO X7$ DR = .4965$
27 GOTO LOOP5$ X7.. DR = 2DR$ GOTO LOOP5$
28 X6.. PBAR = W/173.5$ WRITE('PBAR', PBAR)$
29 FINISH

```

PBAR

1.0842,-01

Fig. 20 -- Program for $\bar{P}_{spn}$.

ALG EFNCY $\bar{P}$
ALGOL spf

MARCH 21, 1965

INTERFACE FEBRUARY 15, 1965

PASS2

```

1  REAL DR, R, W, V, DZ, Z, Q, T, DRP, RP, DTP, S, F, P, TP, PRAR$
   LEVEL 1
2  INTEGER N,M,L,K$
3  N = 1$ DR = 1.27$ R = 13.965$ W = 0$
4  100P5.. M = 1$ V = 0$ DZ = .3310$ Z = .1655$
5  100P4.. Q = 0$ T = R$ DRP = .2 $ RP = R + .1$
6  100P3.. I = 1$ LOOP2.. K = 1$ DTP = .0995$
7  S = SQRT((7.62 - Z)*(7.62 - Z) + R*R + RP*RP - 2*R*RP)$
8  F = R*(7.62 - Z)*RP*(EXP(-.734*S))/(S*S*S)$
9  P = .0645F$ 100P1.. K = 2K + 1$ TP = .0332K$
10 S = SQRT((7.62 - Z)*(7.62 - Z) + R*R + RP*RP - 2*R*RP*COS(TP))$
11 F = R*(7.62 - Z)*RP*(EXP(-.734*S))/(S*S*S)$
12 P = P + F*DTP$ DTP = 2DTP$ IF TP LSS 2 THEN GOTO LOOP1$
13 IF RP LSS R THEN GOTO X1$ L = 2*L + 1$ RP = R + .1L$
14 T = T + DRP$ IF T GTR 14.6 THEN GOTO X20$ Q = Q + P*DRP$
15 IF DRP GTR .25 THEN GOTO X21$ DRP = .3$ GOTO LOOP2$
16 X21.. DRP = 2*DRP$ GOTO LOOP2$ X20.. DRP = DRP - T + 14.6$
17 Q = Q + P*DRP$ T = 0$ DRP = .2$ RP = R - .1$
18 GOTO LOOP3$ X1.. L = 2*L + 1$ RP = R - .1L$ T = T + DRP$
19 IF T GTR R THEN GOTO X2$ Q = Q + P*DRP$
20 IF DRP GTR .25 THEN GOTO X3$ DRP = .3$ GOTO LOOP2$
21 X3.. DRP = 2*DRP$ GOTO LOOP2$ X2.. DRP = DRP - T + R$
22 Q = Q + P*DRP$ V = V + Q*DZ$ IF M GTR 8 THEN GOTO X5$
23 M = 2*M + 1$ Z = .1655*M$ IF DZ GTR .4 THEN GOTO X4$
24 DZ = .993$ GOTO LOOP4$ X4.. DZ = 2*DZ$ GOTO LOOP4$
25 X5.. W = W + V*DR$ IF N GTR 8 THEN GOTO X6$ N = 2N + 1$
26 R = 14.6 - .635*N$ IF DR GTR 1.8 THEN GOTO X7$ DR = 1.905$
27 GOTO LOOP5$ X7.. DR = 2DR$ GOTO LOOP5$
28 X6.. PRAR = W/2550$ WRITE('PRAR', PRAR)$ PRAR
29 FINISH

```

3.0236,-03

-70-

Fig. 21 -- Program for $\bar{P}_{\text{spf}}$

ALG EFFCY $\bar{P}_{LPN}$
ALGOL

MARCH 21, 1965

INTERFACE FEBRUARY 15, 1965

PASS2

```

1  DEAL DR, R, W, V, DZ, Z, Q, T, DRP, RP, DTP, S, F, P, TP, PBAR$
  LEVEL 1
2  INTEGER N, M, L, K$
3  N = 1$ DR = 1.27$ R = 13.965$ W = 0$
4  100P5.. M = 1$ V = 0$ DZ = .662$ Z = .331$
5  100P4.. Q = 0$ T = R$ DRP = .2 $ RP = R + .1$
6  100P3.. L = 1$ 100P2.. K = 1$ DTP = .0995$
7  S = SORT(Z*Z + R*R + RP*RP - 2*R*RP)$
8  F = R*Z*RP*(EXP(-.734*S))/(S*S*S)$
9  P = .0645F$ 100P1.. K = 2K + 1$ TP = .0332K$
10 S = SORT(Z*Z + R*R + RP*RP - 2*R*RP*COS(TP))$
11 F = R*Z*RP*(EXP(-.734*S))/(S*S*S)$
12 P = P + F*DTP$ DTP = 2DTP$ IF TP LSS 2 THEN GOTO 100P1$
13 IF RP LSS R THEN GOTO X1$ L = 2*L + 1$ RP = R + .1L$
14 T = T + DRP$ IF T GTR 14.6 THEN GOTO X20$ Q = Q + P*DRP$
15 IF DRP GTR .25 THEN GOTO X21$ DRP = .3$ GOTO 100P2$
16 X21.. DRP = 2*DRP$ GOTO 100P2$ X20.. DRP = DRP - T + 14.6$
17 Q = Q + P*DRP$ T = 0$ DRP = .2$ RP = R - .1$
18 GOTO 100P3$ X1.. L = 2*L + 1$ RP = R - .1L$ T = T + DRP$
19 IF T GTR R THEN GOTO X2$ Q = Q + P*DRP$
20 IF DRP GTR .25 THEN GOTO X3$ DRP = .3$ GOTO 100P2$
21 X3.. DRP = 2*DRP$ GOTO 100P2$ X2.. DRP = DRP - T + R$
22 Q = Q + P*DRP$ V = V + Q*DZ$ IF M GTR 8 THEN GOTO X5$
23 M = 2*M + 1$ Z = 7.62 - .331*M$ IF DZ GTR .8 THEN GOTO X4$
24 DZ = .993$ GOTO 100P4$ X4.. DZ = 2*DZ$ GOTO 100P4$
25 X5.. W = W + V*DR$ IF N GTR 8 THEN GOTO X6$ N = 2N + 1$
26 R = 14.6 - .635*N$ IF DR GTR 1.8 THEN GOTO X7$ DR = 1.905$
27 GOTO 100P5$ X7.. DR = 2DR$ GOTO 100P5$
28 X6.. PBAR = W/2550$ WRITE('PBAR', PBAR)$ PBAR
29 FINISH

```

1.2583, -01

Fig. 22 -- Program for $\bar{P}_{lpn}$.