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New Application of Delayed Coincidence Techniques for Measuring Lifetimes of Excited Nuclear States—Ca^{42} and Sc^{47}

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A new application of the delayed coincidence technique for measuring short nuclear lifetimes has been developed which is basically similar to the well-known prompt comparison method. Systematic errors can be reduced and greater accuracy obtained for nuclei which exhibit both a prompt and a delayed event. A transistorized time-to-amplitude converter was used. The mean life of the second excited state of Ca^{42} $[(4.8\pm0.3)\times10^{-10}$ sec$]$ and the first excited state of Sc^{47} ($<5\times10^{-12}$ sec) were measured. Also the mean life of the first excited state of Hg^{198} $[(3.5\pm0.5)\times10^{-11}$ sec$]$ was determined by the usual self-comparison method.

I. INTRODUCTION

For many years delayed coincidence techniques have been used to measure short nuclear lifetimes.[1] Originally coincidence counting rates were observed as a function of delays inserted with mechanical delay lines or cables. Now electronic time-to-amplitude converters are generally employed with multichannel analyzers to increase greatly the efficiency of data collection.[2] In either case, the data analysis is quite similar. Some lifetimes can be determined from the exponential slope of the cable or amplitude distribution curves. Shorter lifetimes can be measured by analysis of the centroid shift of these distributions.[3]

The direct observation of an exponential slope is certainly the simplest and least ambiguous approach, but this method is limited by present experimental techniques to lifetimes of the order of 10^{-10} sec or longer. Centroid analysis can reduce this lower limit by another order of magnitude but additional complications are introduced. In order to measure lifetimes by centroid shift, it is necessary to determine "time-zero"—i.e., the centroid of the time distribution for a prompt coincidence event. Usually a comparison is made with a second source which exhibits a prompt event between particles similar to the delayed event being studied. Great care must be taken in generating the prompt distribution to insure that it is a valid comparison for the delayed event under consideration. One method of circumventing the necessity for a second source is the self-comparison technique for beta-conversion electron coincidences.[1] This method is powerful for a limited number of cases and has been used during the course of the present work with an accurady of 5×10^{-12} sec. The technique, however, requires a double coincidence magnetic spectrometer—an instrument not generally available in every laboratory.

When it is necessary to use a second source for prompt comparison studies, it is inevitable that systematic errors are going to be introduced. The two sources must be interchanged frequently to minimize drifts in the equipment. They must be positioned very accurately because small changes in flight paths represent relatively large time delays, and of course the particles emitted by both sources must be similar. In order to minimize the systematic errors normally present in the prompt comparison technique, the present work has concentrated on nuclei where a single source exhibits an interesting delayed event as well as a prompt comparison event.

II. PRINCIPLE OF THE METHOD

There are two important conditions which must be met in order to apply this new technique. First, a source is needed which exhibits the delayed coincidence event to be measured and a prompt coincidence event to be used for comparison.[4] These two events must *not* be distinguishable in a time-to-amplitude converter. Second, the two events must be distinguishable by an auxiliary coincidence system.

The prompt and delayed coincidence events are observed simultaneously by two fast-plastic scintillation detectors connected to a time-to-amplitude converter. The output pulses from the time converter are fed into a 256-channel analyzer for storage. There are many isotopes which are suitable because they exhibit both a prompt and a delayed event. However, it is not necessary that both events be present in one isotope. Two isotopes can be mixed to form a single source. The important consideration is that the time converter must respond identically to both events. This of course requires that the initial particles in both events and the

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[1] R. E. Bell, R. L. Graham, and H. E. Petch, Can. J. Phys. **30**, 35 (1952).

[2] R. E. Green and R. E. Bell, Nuclear Instr. **3**, 127 (1958).

[3] Z. Bay, Phys. Rev. **77**, 419 (1950).

[4] The term "coincidence event" used here refers to the emission of one particle when an excited state is formed and a second particle when the state decays. The second particle is delayed on the average by the mean life of the excited state. When the mean life is very short ($\leq10^{-12}$ sec), the event is considered "prompt," i.e., the electronic system cannot detect any time delay between the two particles. When the mean life is longer ($\geq5\times10^{-12}$ sec), it is possible in many cases to measure the delay electronically. These events are then referred to as "delayed" coincidences.

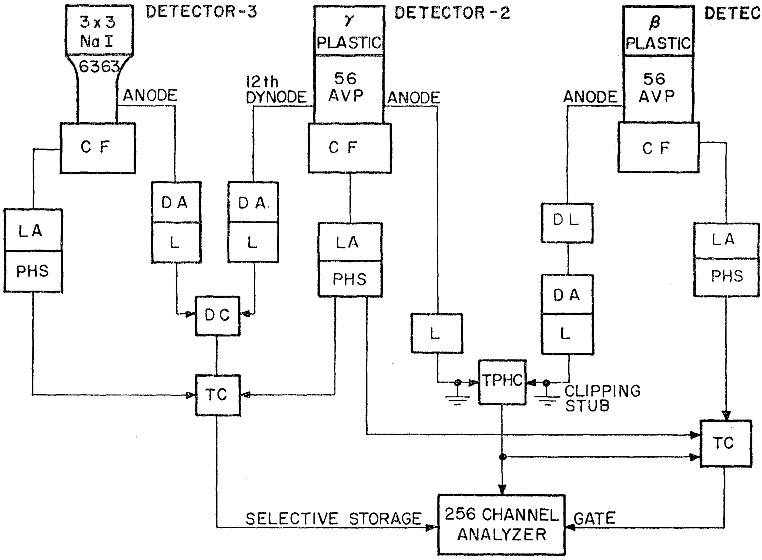

FIG. 1. Electronics block diagram. *CF* = cathode follower, *DA* = distributed amplifier, *LA* = linear amplifier, *L* = limiter, *PHS* = pulse-height selector, *DL* = 50-ohm delay line, *TPC* = time to pulse height converter, *DC* = double coincidence, *TC* = triple coincidences.

final particles in both events lose the same amount of energy in the plastic scintillators.

In order for an event to be distinguishable, a third particle must be emitted in coincidence with the two particles which are observed by the fast scintillation detectors. A third detector operated in coincidence with one of the two fast detectors is used to recognize the event. A complete distinction can be made when both the prompt and delayed events exhibit a triple coincidence. However, in many cases it is not necessary to completely distinguish both types of events. For example, suppose that the prompt events occur much more frequently than the delayed events. Then most of the pulses sent to the analyzer by the time converter would be characteristic of the prompt event; and even if the delayed events were ignored, the observed time distribution can be used to define time-zero to a good approximation. The third detector now comes into play. With it, a fraction of the delayed events can be distinguished and stored concurrently with the prompt events but in a separate part of the analyzer memory. The average delay of the delayed event can then be determined by comparing the centroids of the prompt and delayed time distributions. The method is quite useful even if the prompt distribution is slightly distorted by the presence of some delayed events. The distortion is only a fraction of the delay time being measured. That is, for very short delays where greater accuracy is needed, the distortion is small.

This short discussion has only considered the basic principles of the technique. The method will work for any case where a prompt and a delayed event are present which are not distinguishable in the fast channels but are distinguishable by additional detectors. Systematic errors are greately reduced because the prompt and delayed time distributions are recorded concurrently by the multichannel analyzer. Two specific examples will be discussed subsequently, and more of the experimental problems will then be considered.

III. EXPERIMENTAL ARRANGEMENT

A block diagram of the electronics system is shown in Fig. 1. Detectors one and two (*D*1 and *D*2), followed by appropriate shaping circuits, provide the pulses for the time-to-amplitude converter. Fast plastic scintillators are used for these detectors in order to reduce the time jitter of the detection process. The pulses from the time converter are fed into a 256-channel analyzer which is gated by a triple coincidence. This part of the system is quite conventional, and the prompt and delayed events could not be distinguished without the assistance of a third channel.

The third channel uses a 3×3 NaI detector (*D*3) operated in coincidence with *D*2 by the usual fast-slow arrangement. The output of this circuit is used to control a selective storage system in the 256-channel analyzer. If there is no $D2-D3$ coincidence, the pulse representing timing information is stored in the lower half of the analyzer (i.e., channels 1–128). But when there is a $D2-D3$ coincidence, declaring that a delayed event has occurred, the associated time pulse is stored in the upper half of the analyzer (channels 129–256).

An air-core coaxial delay line built in the form of a trombone slide was used for time calibration (General Radio Type 874-LT 50 ohms). The slide was mounted with a gear mechanism so that delays as short as 10^{-11} sec could be easily inserted. The most important circuit in the electronic system is a newly designed transistorized time-to-amplitude converter.[5] It is a very simple, reliable circuit using only three transistors. The

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[5] P. C. Simms (to be published). The circuit used in this work as well as an improved model will be described in detail. Application of the same transistor configuration to other fast coincidence requirements will also be discussed.

performance of the instrument was not optimized in the present work because only the 50-ohm delay line was available. If a higher impedance delay line had been available, the time resolution could have been improved by removing the distributed amplifier in channel 1. The circuit performed satisfactorily even with single counting rates as high as 200 000 counts per second. Double delay line clipped linear amplifiers were used to handle these high counting rates.

IV. PERFORMANCE

A prompt resolution curve for the time-to-amplitude converter is shown in Fig. 2. The full width of the curve at half maximum is 5×10^{-10} sec, and the slope for a reduction of counting rate by a factor of two is 10^{-10} sec. The delay line was calibrated by comparing delay time to gamma-ray time-of-flight and by observing standing waves in the line with a radio-frequency impedance bridge. The velocity of an electrical pulse down the line was found to be equal to the velocity of light with an accuracy of better than 3%.

The performance and accuracy of the time converter was checked by measuring the well-known mean life of the first excited state of Hg^{198}. The self-comparison technique was employed with beta-conversion electron coincidences in a Gerholm double coincidence magnetic spectrometer. The result obtained, $\tau=(3.5\pm0.5)\times10^{-11}$ sec, is in good agreement with previous results which have been determined by several different techniques.[6]

The selective storage part of the system was checked by using annihilation quanta from a Na^{22} source. The two plastic detectors were positioned at an angle of 180°. Each detector (size $1\frac{1}{2}$ in.$\times1\frac{1}{2}$ in.) was one inch from the source. Considering the solid angles subtended and the difference in efficiency for detecting the 1.28-Mev γ ray and the 0.511-Mev γ ray, it was estimated that the probability for detecting a 1.28–0.511 coincidence was less than 0.5% of the probability for detecting a coincidence between the two annihilation quanta. Thus, practically all of the time pulses represent prompt coincidences. There is a delay of the order of 2×10^{-10} sec between the 1.28- and the 0.511-Mev γ rays. However, since the probability for detecting the 1.28-Mev γ rays is essentially the same for both detectors, the small number of 1.28–0.511 coincidences present could at most only slightly broaden the time distribution. These events could not create a net shift in the centroid of the distribution. The selective storage system was operated by the 1.28-Mev γ ray in detector 3. Most of the time pulses were stored in the lower half of the analyzer because the probability for detecting the 1.28-Mev γ ray in $D3$ was relatively small. When the triple coincidence was observed, the associated time pulse was stored in the upper half of the analyzer. As expected, the mean value of the two distributions was found to be the same with a statistical accuracy of 10^{-12} sec.

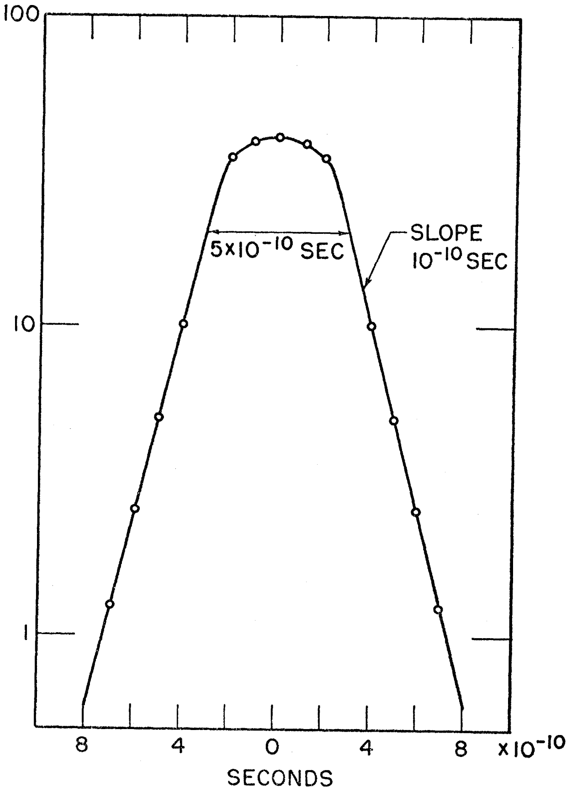

FIG. 2. Prompt resolution curve with Co^{60}.

V. INVESTIGATION OF Ca^{42} AND Sc^{47}

The second excited state of Ca^{42} was selected for this work because of the theoretical interest in 0^+ excited states. It has been pointed out by Church and Weneser[7] that accurate determination of transition probabilities for electric monopole conversion electrons would yield information about nuclear structure. In addition to the lifetime of the 0^+ state, one needs to know the branching ratios for the various modes of decay of the state. These branching ratios have been measured recently and the results for the nuclear "strength parameter" ρ will be discussed in a subsequent paper.[8]

The decay scheme of K^{42} is shown in Fig. 3. The lifetime of the first excited state is very short ($\sim10^{-13}$ sec) because of the large transition energy (1.53 Mev).

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[6] $(3.15\pm0.3)\times10^{-11}$ sec, resonance fluorescence: F. R. Metzger and W. B. Todd, Phys. Rev. **95**, 853 (1954). $(3.2\pm0.7)\times10^{-11}$ sec, resonance fluorescence: W. G. Davey and P. B. Moon, Proc. Phys. Soc. (London) **A66**, 956 (1953). 3×10^{-11} sec, Coulomb excitation: R. Barloutaud, T. Grjebine, and M. Riou, Physica **22**, 1129 (1956). $(3\pm1)\times10^{-11}$ sec, delayed coincidence: A. W. Sunyar, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, 1958* (United Nations, Geneva, 1958).

[7] E. L. Church and J. Weneser, Phys. Rev. **103**, 1035 (1956).

[8] N. Benczer-Koller, M. Nessin, and T. H. Kruse, Bull. Am. Phys. Soc. **5**, 248 (1960).

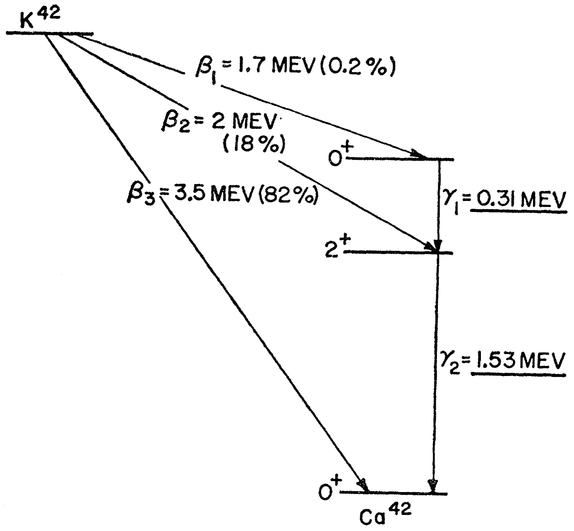

FIG. 3. Decay scheme of K^{42}.

Thus a $\beta_2-\gamma_2$ coincidence will be a "prompt" event which can be used to mark time-zero. A $\beta_1-\gamma_2$ coincidence will be a delayed event, i.e., delayed on the average by the mean life of the second excited state. The delayed event can be distinguished by detecting a coincidence between γ_1 and γ_2.

The source-detector arrangement used in investigating Ca^{42} is shown in Fig. 4. In a triple coincidence experiment it is important that the detectors subtend as large a solid angle as possible. Still the gamma detectors cannot be placed arbitrarily close to the source since they must be shielded from beta particles. In order to stop the high-energy beta particles without creating bremsstrahlung, a thick low Z material must be used. The beta crystal was thick enough ($\sim$1 cm) to stop the beta particles of interest and yet thin enough to minimize the probability for gamma detection. The plastic gamma detector size was selected as a compromise between detection efficiency and time-jitter for light collection from the crystal.

The plastic beta detector ($D1$) was set to detect beta particles with energies around 1 Mev. Thus it could detect either β_1 or β_2. The other plastic detector ($D2$), which was shielded from betas, was used to detect γ_2; and the 3×3 detector was used to detect the low intensity γ_1. When a $\beta-\gamma$ coincidence was observed by $D1$ and $D2$, a pulse was sent by the time converter to the 256-channel analyzer. If there was no $\gamma_1-\gamma_2$ coincidence, the pulse representing timing information was stored in the lower half of the analyzer (i.e., channels 1–128). But when there was a $\gamma_1-\gamma_2$ coincidence, declaring that the event observed in $D1$ and $D2$ was a delayed event, the associated time pulse was stored in the upper half of the analyzer (channels 129–256). Since the efficiency for detecting γ_1 was only $\sim$10% most of the delayed events were missed and stored incorrectly with the prompt events in the lower half of the analyzer. Nevertheless, there was no appreciable distortion of the prompt distribution because the relative number of delayed events was so small. Then by comparing the centroid of the delayed distribution to the centroid of the prompt distribution, the lifetime of the second excited state was determined.

Before this work, there was no information to indicate whether the first excited state of Sc^{47} (see Fig. 5) had an energy of 0.82 or 0.49 Mev. Thus the object was to see if either the $\beta_1-0.49\ \gamma$ event or the $\beta_1-0.82\ \gamma$ event was delayed. If so, the lifetime of the first excited state could be measured and the order of the gamma rays determined. The same experimental arrangement was used as for Ca^{42}. Channel 1 was set to detect electrons with energies of $\sim$0.2 Mev. The pulse-height selector in channel 2 was set on the Compton distribution of γ_3 and the selector in channel 3 on the γ_2 photopeak. In this way detector 2 can detect either γ_1 or γ_3 (or even γ_2 if γ_2 has higher energy than γ_3). A $\beta_1-\gamma_1$ or γ_2 coincidence is a "prompt" event while a $\beta_1-\gamma_3$ event is "delayed." The delayed events are distinguished by a $\gamma_2-\gamma_3$ coincidence which operates the selective storage circuit. In this case it is obvious that an appreciable number of delayed events can be mixed into the prompt time-distribution, but the energies of the three gamma rays are distinct enough that the delayed time-distribution should be pure. The relative probability of detecting γ_1 or γ_3 in $D2$ controls the amount of mixing in the prompt distribution. The mixing can be determined and corrected for, or the amount of mixing can be varied by the pulse-height selector in channel 2 and the correct result obtained by extrapolation.

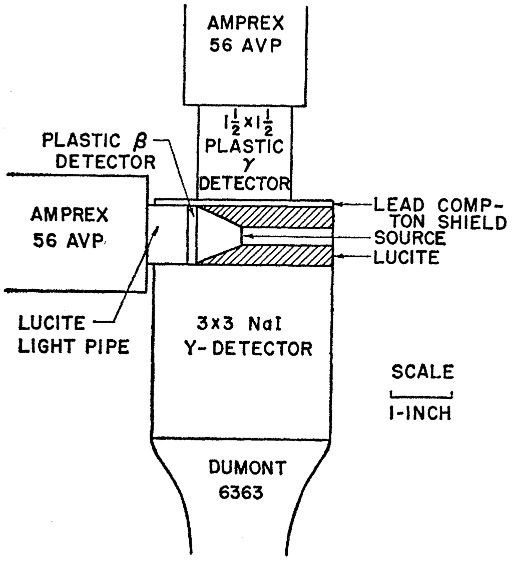

FIG. 4. Detector-source arrangement.

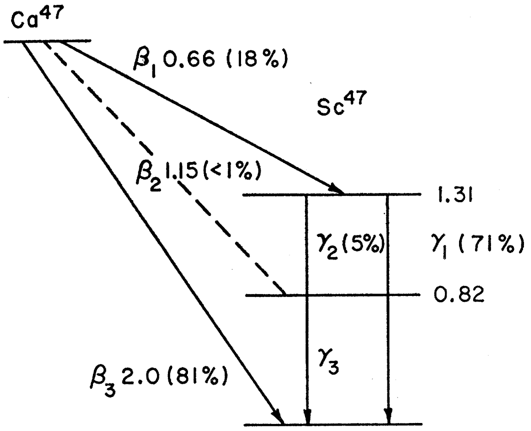

FIG. 5. Decay scheme of Ca^{47}.

VI. ANALYSIS OF RESULTS

In order to understand the accuracy of the results that can be obtained with this method, the role of the third detector must be carefully considered. Its addition to the standard two-channel time converter would not be justified merely to distinguish the delayed events from a background of prompt events. The rate of accumulation of the triple coincidences is so small that the stability of this sensitive instrument would be inadequate for maximum accuracy if it were necessary to alternate periodically between prompt and delayed coincidences. In the arrangement described here, where the prompt and delayed events are recorded concurrently, there are plenty of prompt events (since only a double coincidence is required) which can be used to monitor instrumental instabilities and define time-zero.

One must then question what possible differences exist between the prompt and delayed events. No major problem is presented in insuring that the same amount of energy is lost in the two fast detectors for the two types of events. In some cases small single-channel analyzer window widths must be used to minimize differences in the spectral distribution of the various particles. Instrumental drifts and instabilities—such as those due to counting rate changes—are certainly minimized since the events are recorded concurrently. There is no problem with the location of two sources. The operation of the selective storage system in the multichannel analyzer is direct and simple. All pulses are processed in the same manner by the analog-to-digital converter. The two events are separated at the address scaler. Normally the address scaler is reset to zero before a channel is assigned to a particular pulse. The selective storage system merely resets the address scaler to 128 rather than to zero when the delayed event is identified. Incidentally, multichannel analyzers which do not include this feature may be easily modified to provide the selective storage function.

It would seem then that the processing of the time-sensitive information is quite reliable and the only major problem that exists is distinguishing between the delayed and prompt events with the third channel. This was found to be somewhat difficult in the Ca^{42} experiment due to the presence of Compton scattering from detector 2 to detector 3. (See Fig. 4) When the 1.53-Mev γ ray is recorded in detector 2, the energy lost in the plastic scintillator is from a Compton scattered electron. If the incident γ ray loses a large portion of its energy in the crystal, the scattered γ ray will have low energy and be directed backward from the incident γ ray. The effect of the scattered γ ray can then be quite serious since γ_1 has low energy and low intensity.

If the 1.53-Mev γ ray is recorded in detector 2 and then scattered and recorded in detector 3, a prompt event ($\beta_2-\gamma_2$) will be falsely identified as a delayed event. Thus the delayed distribution could be badly distorted. Lowering the acceptance window in channel 2 helped to reduce the effect, but it was still absolutely necessary to include the lead Compton shield shown in Fig. 4 in order to be able to distinguish the 0.31-Mev γ ray clearly. (See Fig. 6.) The shield was selected ($\frac{3}{16}$ in. thick) to permit most high-energy γ rays to pass through to detector 2 while reducing the Compton scattering back into detector 3. An even better detector arrangement as far as Compton scattering is concerned would have been to place the two γ detectors at a 90° angle. Nevertheless, the 180° position was chosen because of the pronounced $\gamma-\gamma$ directional correlation in a $0-2-0$ cascade.[9] Even when using the shield, the delayed distribution was carefully corrected for the presence of a small number of prompt events.

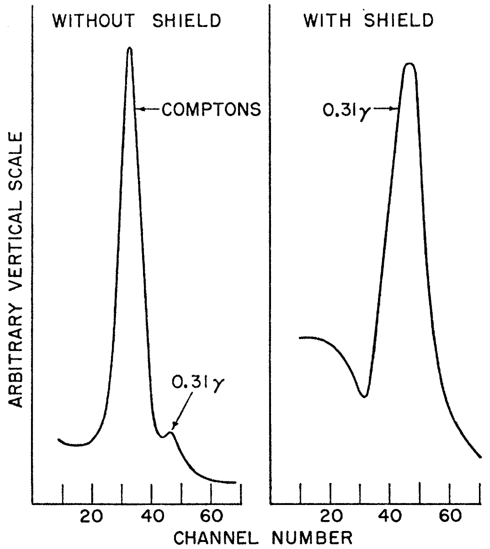

FIG. 6. Gamma spectrum of 3×3 detector in coincidence with detector 2.

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[9] C. S. Wu, Pegram Nuclear Physics Laboratories Progress Report CU (PNPL)-202 (unpublished).

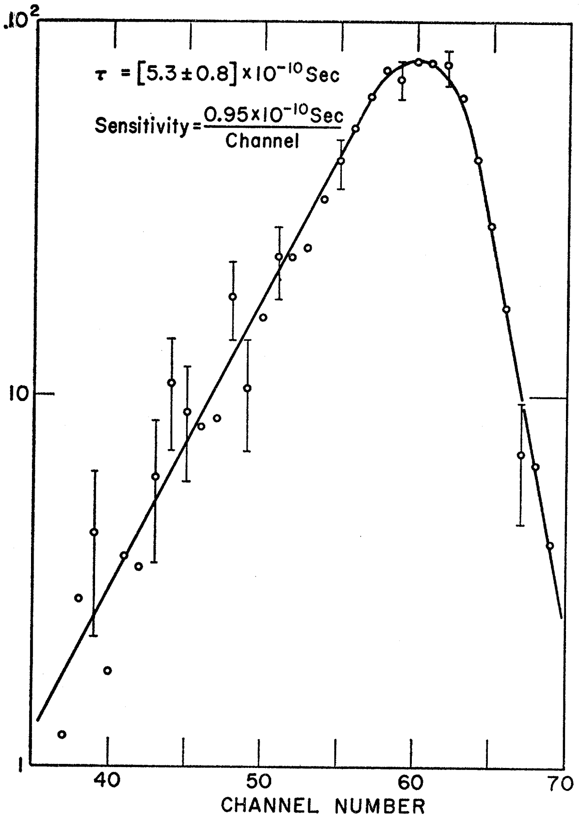

FIG. 7. Delayed time-distribution for Ca^{42}.

The centroids for the timing distributions were calculated from the following formula:

$$\bar{x}=\sum_i y_i x_i / \sum_i y_i,$$

where x_i is the channel number, y_i is the number of counts per channel, and the sum is over all channels. The standard error in the mean value of the distribution is given by[10]

$$(\sigma_{\bar{x}})^2 \simeq \frac{1}{n(n-1)} \sum_1^n (x_i-\bar{x})^2,$$

where n is the total number of counts in the distribution. This can be rewritten more directly in terms of the observed data as

$$(\sigma_{\bar{x}})^2 \simeq \frac{1}{(\sum_i y_i)^2} \sum_i y_i (x_i-\bar{x})^2,$$

which is equivalent to

$$\sigma_{\bar{x}} \simeq \frac{1}{\sum_i y_i} \left[(\sum_i y_i x_i^2) - \bar{x} (\sum_i y_i x_i) \right]^{\frac{1}{2}}.$$

VII. RESULTS AND CONCLUSIONS

The result obtained for the mean life of the second excited state of Ca^{42} was

$$\tau = (4.8 \pm 0.3) \times 10^{-10} \text{ sec}.$$

The limit of error quoted contains both statistical and systematic errors but is primarily due to the uncertainty in correcting for Compton scattering. An additional check was made on the order of magnitude of the result by analyzing the exponential slope of the delayed distribution (Fig. 7). The two results are in good agreement, but the result from the centroid analysis is considered more accurate because a much larger fraction of the data was used in the determination.

In the Sc^{47} experiment it was found that neither the $\beta_1 - 0.49\ \gamma$ or $\beta_1 - 0.82\ \gamma$ events were delayed by a measurable amount. That is, for both cases when either the 0.49 or 0.82 gamma rays were considered to be γ_3, there was no measurable difference in the centroids of the "delayed" and prompt distributions. There was a 20% mixing of "delayed" pulses in the prompt distribution; but since both distributions were in reality prompt, no correction for this mixing was necessary. Thus only an upper limit can be set on the mean life of the first excited state of Sc^{47}:

$$\tau \leq 5 \times 10^{-12} \text{ sec}.$$

Since γ_3 is believed to be produced by an $E2$ transition,[11] this short lifetime strongly indicates that the energy of the level must be 0.82 Mev and not 0.49 Mev. In order to support this assignment, an additional effort was made to observe the β_2 spectrum. The spectrum was not observed, but an improved upper limit on its existence of less than 1% was established.

ACKNOWLEDGMENTS

The authors would like to acknowledge the assistance of Mr. Luke Mo in running the experiment and Mr. Jack Hahn and his electronics staff in the preparation of the equipment.

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[10] See, for example, R. D. Evans, *The Atomic Nucleus* (McGraw-Hill Book Company, New York, 1955), Chap. 26.

[11] L. J. Lidofsky and V. K. Fischer, Phys. Rev. **104**, 759 (1956).