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Directional correlation of the 346-136 keV gamma-gamma cascade in Ta¹⁸¹

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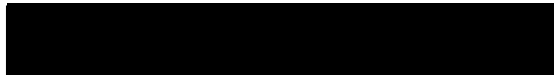
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AN ABSTRACT OF THE THESIS OF Robert Allan Wilson for the
Master of Science in Physics presented May 23, 1969.


Title: Directional Correlation of the 346-136 Kev Gamma-Gamma
Cascade in Ta¹⁸¹.

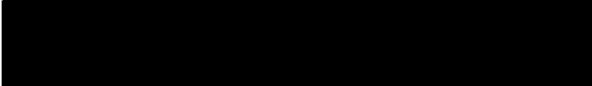
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Donald Wright

The directional correlation of the 346-136 Kev gamma-gamma cascade in Ta¹⁸¹ was measured using the delayed coincidence method with a source of Hf¹⁸¹ in 27 N HF. The contributions to the composite delayed coincidence spectrum of the interfering 133-482 Kev and 133-346 Kev cascades were removed by the subtraction

from this spectrum of appropriate 133-482 Kev pure delayed coincidence data. The extracted 346-136 Kev coincidence data then yielded the correlation coefficients $A_{22} = 0.190 \pm 0.011$ and $A_{44} = -0.025 \pm 0.024$ that are consistent with the established spin sequence $5/2(E2)9/2(M1 + E2)7/2$ and with an admixture of $16.2\% \pm 1.2\%$ E2 radiation in the 136 Kev mixed transition. The results of the experiment are in excellent agreement with those obtained from conversion electron measurements and resolve the discrepancy apparent in earlier work on the 346-136 Kev cascade.

DIRECTIONAL CORRELATION OF THE 346-136 Kev
GAMMA-GAMMA CASCADE IN Ta¹⁸¹

by

ROBERT ALLAN WILSON

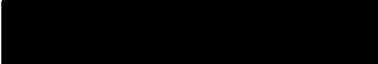
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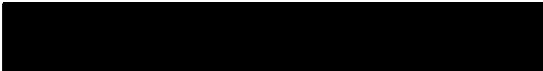
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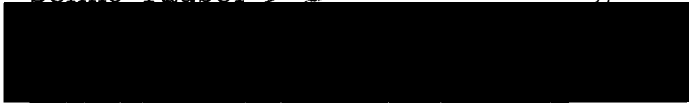
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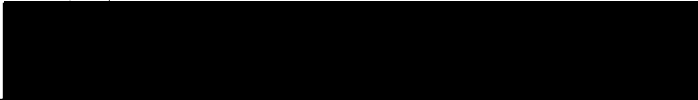

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INTRODUCTION

In the field of low-energy nuclear physics, the low-lying excited level structure of nuclei is of primary interest and it is the business of nuclear spectroscopy to obtain detailed information on this level structure through the investigation of nuclear properties. Among these properties are the spin angular momenta of excited states and the multipole orders and relative multipole composition of radiative transitions between these states. A powerful experimental method often used to evaluate these nuclear properties is the directional correlation measurement where, for a nucleus decaying by the emission of two particles or radiations in cascade (i. e. in rapid succession), the directional distribution in space of the second particle or radiation relative to the first is obtained. Despite practical limitations restricting its usefulness, the directional correlation technique has been instrumental as an aid in the establishment of consistent decay schemes for many nuclei (1). Over the last decade the growth in sophistication of nuclear instrumentation has led to the application of this technique to increasingly difficult experimental problems, and to the refinement of measurements obtained in earlier work as well. This thesis affords an example

of the latter, where a re-determination of earlier measurements yielded significantly different results.

A partial decay scheme (1, p. 6-6-138) for the Ta^{181} nucleus is shown in Figure 1 where the 346 Kev-136 Kev gamma-ray cascade, the subject of the current work, is emphasized. Although this cascade has been investigated by McGowan (2) in 1953 and later by Debrunner et al. (3) in 1956, the results of these two studies are not in agreement. A directional correlation measurement of this cascade is complicated by the unavoidable collection of data from interfering cascades present in this nucleus and by the resulting necessity of distinguishing this unwanted data in some way from the desired data. It was thought that, with the use of modern apparatus, a carefully conducted directional correlation measurement of the 346-136 Kev cascade could overcome these experimental difficulties and resolve the discrepancy in the earlier work. The resolution of this discrepancy should then provide an additional verification of the established spin sequence $5/2(E2)9/2(M1 + E2)7/2$ characterizing the transitions and excited states emphasized in Figure 1. In addition to this possibility, the performance of such a measurement was expected to provide an operational test of the apparatus and to develop procedures and techniques useful in future work. It was to attain these objectives that the experiment to be described in this thesis was undertaken.

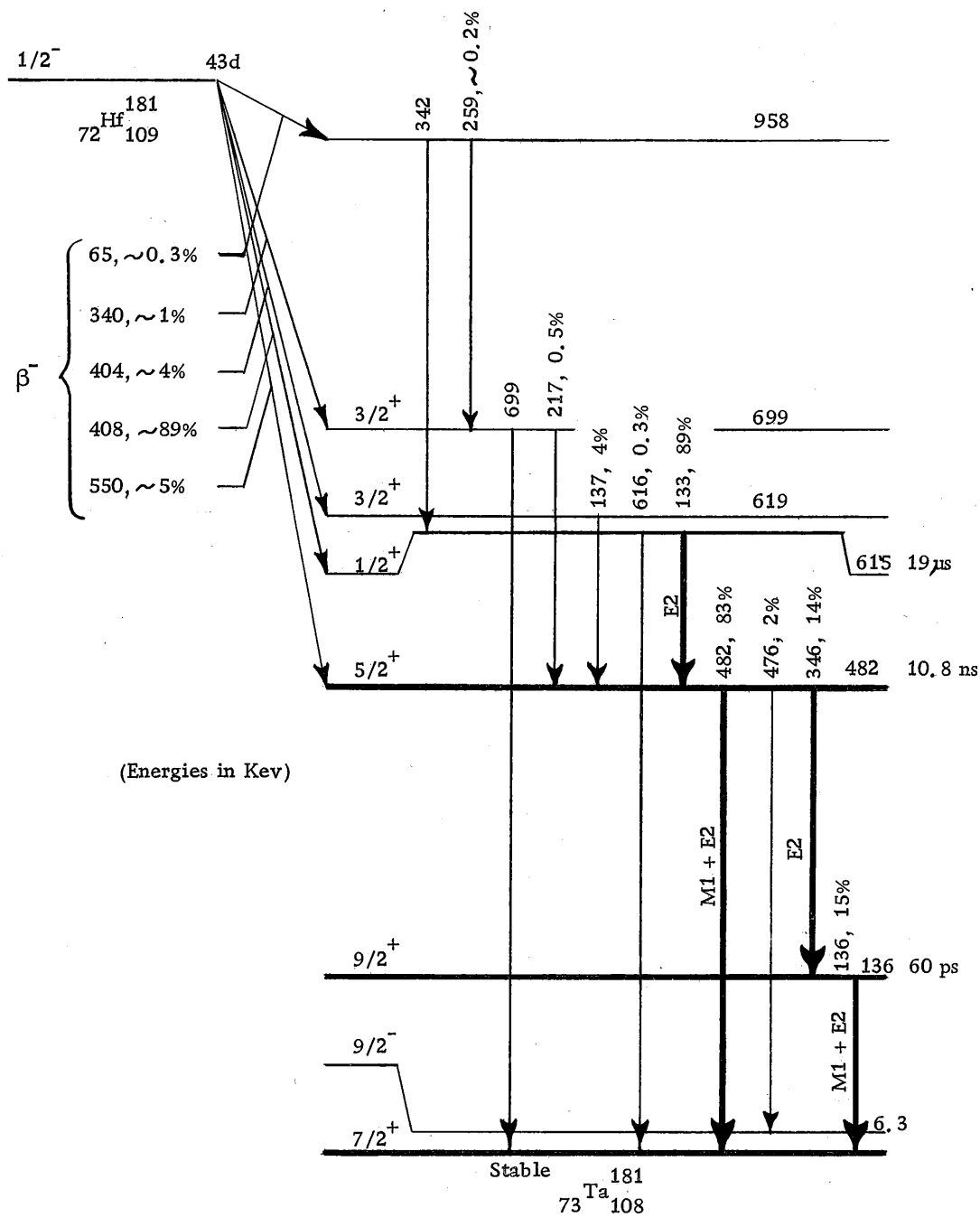


Figure 1. Partial decay scheme in Ta^{181}

THEORY

The general theory of the directional correlation is complete and is presented in detail in the literature (4, 5). Only the results of the theory pertinent to the experiment will be presented, any more extensive treatment being beyond the scope of this thesis. Nevertheless, a simple phenomenological discussion of the basic directional correlation process will serve to define terms, introduce the pertinent notation and provide some insight into the correlation mechanism.

In directional correlation work, a typical situation is shown in Figure 2. a, where a nucleus in an excited state a, decays to a short-lived intermediate excited state b, and then to a state of lower energy c. Two gamma rays whose energies are equal to the differences in energy of the states are emitted in cascade in rapid succession. These gamma rays are characterized by angular momentum quantum numbers L, projection quantum numbers M and propagation vectors \vec{k} . The excited states have spin quantum numbers I and quantum numbers m that are projections of the I onto an arbitrary quantization axis. The transitions among these excited states are governed by the selection rules shown below the figure.

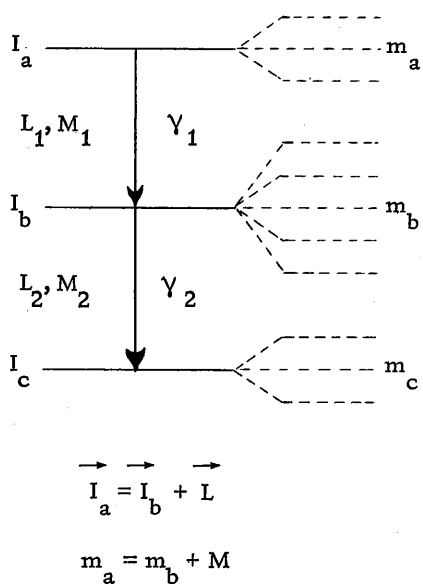


Figure 2. a. The Cascade.

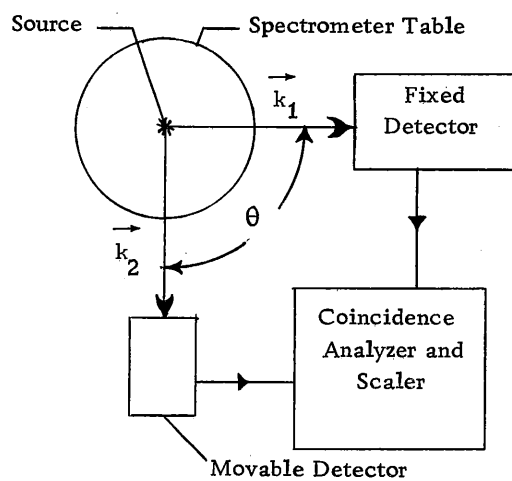


Figure 2. b. The Apparatus.

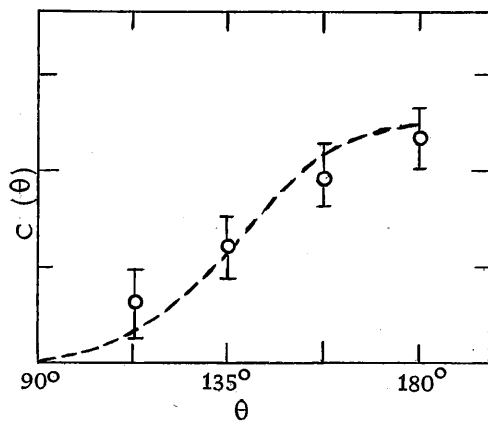


Figure 2. c. The Result.

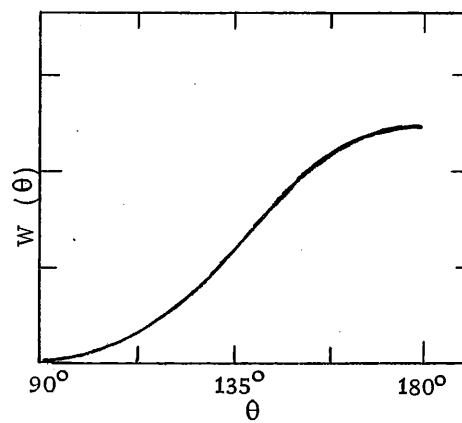


Figure 2. d. The Theory.

Figure 2. The directional correlation experiment

A very simple apparatus for measuring the directional correlation of the cascade is shown in Figure 2. b. The detectors are sensitive to gamma rays emitted by a source containing a large number of nuclei that decay in the manner previously described. The coincidence circuit allows only gamma-ray pairs emanating from the same nucleus to be counted by the scaler. A typical coincidence count rate thus obtained might appear as shown in Figure 2. c and is often found to be non-isotropic.

From the data in Figure 2. c a directional correlation function $W(\theta)$ can be obtained and this function is the relative probability that γ_2 will be emitted into the increment of solid angle $d\Omega$ at an angle θ with respect to \vec{k}_1 . The most common theoretical expression for the interpretation of experimental results is the series (6; 5, p. 1029-1031)

$$W(\theta) = 1 + \sum_{k \text{ even}} A_{kk} P_k(\cos \theta) \quad (1)$$

where $0 \leq k \text{ even} \leq \text{smallest of the integers } 2I_b, 2L_1, 2L_2$.

The coefficients A_{kk} are products of coefficients each of which depends only on the properties of a particular transition of the cascade. That is,

$$A_{kk} = A_k^{(1)} (L_1 L_1' I_a I_b) A_k^{(2)} (L_2 L_2' I_b I_c). \quad (2)$$

When two multipole components L and L' contribute to a single gamma ray, the radiation is said to be mixed. In this case, the coefficient $A_k^{(1)}$ in equation (2) is given by the expression

$$A_k^{(1)}(L_1 L'_1 I_a I_b) = \frac{A_k(L_1 L_1 I_a I_b) + 2\delta_1 A_k(L_1 L'_1 I_a I_b) + \delta_1^2 A_k(L'_1 L'_1 I_a I_b)}{1 + \delta_1^2} \quad (3)$$

with a similar expression for $A_k^{(2)}$. The mixing ratio δ is defined by the expression (5, p. 1035)

$$\delta^2 = \frac{\text{Int}(L')}{\text{Int}(L)}, \quad (4)$$

where $\text{Int}(L')$ and $\text{Int}(L)$ are the total (i. e. angle integrated) intensities of the L' and L components. The A_k coefficients have been calculated for most cases of interest and are compiled by Ferentz and Rosenzweig (7; 5, p. 1687).

In a typical experiment, the A_{kk} are extracted from a least-squares fit of the function $W(\theta)$ as given by equation (1) to the experimental data. Comparison of these experimental A_{kk} with the tabulated coefficients of equation (2) then yields information on the spins, multipole orders and mixing ratios characterizing the cascade. Such a technique can then contribute to an unambiguous determination of these nuclear parameters.

The question as to why a directional correlation should be observed can be answered by the following argument. Consider first

only the gamma transitions L_1 . The probability of emission of a gamma ray in a particular transition $m_a \rightarrow m_b$ is a function of the angle between the propagation direction of the gamma ray and the nuclear spin axis and is, in general, non-isotropic. However, at room temperature and in the absence of strong magnetic fields the substates m_a are equally populated and thus all transitions from these magnetic substates are equally probable. Because scintillation detectors cannot distinguish among the L_1 gamma rays of different $M = m_a - m_b$ value, the observed radiation will be an average over these non-isotropic components and will be isotropic.

A directional dependence of the gamma rays can be observed only if by some means the population of the substates m can be made non-uniform. It is just this condition that is effected by the coincidence requirement. To see this, one must now consider the entire cascade. All photons detected by the fixed detector will have propagation vectors parallel to an axis containing source and detector. Let this axis be chosen as the quantization axis with respect to which the M quantum numbers are defined. Then, since a photon can transport only its intrinsic angular momentum \hbar along its direction of propagation, only $M = \pm 1$ gamma-ray photons will be observed by the fixed detector. Since only a nucleus decaying with an $M = \pm 1$ transition to a substate m_b can produce such radiation, only such a nucleus, decaying from the same substate m_b can emit

a γ_2 satisfying the coincidence requirement. Because only certain substates m_b can be populated by $M = \pm 1$ transitions, the coincidence requirement in effect selects an unequally populated set of substates m_b and therefore γ_2 photons will not in general have an isotropic radiation pattern with respect to \vec{k}_1 .

EXPERIMENT

I. SOURCE PREPARATION

The Hf^{181} activity was produced in the nuclear reaction ${}_{72}^{\text{Hf}^{180}} + {}_0^1\text{n} \rightarrow {}_{72}^{\text{Hf}^{181}}$ and was obtained in the form of hafnium oxychloride dissolved in HCl. The gamma-ray spectrum of this material was found to contain ~ 905 Kev and ~ 1140 Kev gamma rays in coincidence indicating the presence of an impurity. This impurity, assumed to be 84-day Sc^{46} by Borovikov et al. (8), was effectively removed from the liquid by means of an ion-exchange procedure. The purified source material was dried by evaporation, dissolved in 27 N HF and placed in a plastic tube of 3 mm diameter and 7 mm length. The counting source thus prepared had an activity of about 10 microcuries and approximated a point-source for the particular geometry of the experiment.

II. APPARATUS

General Description

The apparatus used in this experiment is designed to allow a study of both the temporal and the spatial relationships between the member radiations of the 346 Kev-136 Kev gamma-gamma cascade

of interest. More specifically, the system measures the time interval between the emission of the member gamma rays of each cascade pair, and records for each time interval the number of such pairs detected. In order to be recorded, the gamma rays of each pair must have the correct energies, must emanate from a common nucleus and must have propagation vectors making the angle θ subtended by the gamma-ray detectors at the source. These detectors were spaced 5.0 cm from the source and consisted of cylindrical 1" X 1½" NaI (Tl) scintillation crystals mounted clear of scattering material and optically coupled directly to Phillips type 56AVP multiplier phototubes.

The entire apparatus is shown schematically in the block diagram of Figure 3 and, without regard to details of the operation of individual components, the system accomplishes the preceding tasks in the following way. When two gamma rays γ_1 and γ_2 interact with the detectors of channels 1 and 2 respectively, two pulses designated "fast" and "slow" are produced in each channel. Processing of the fast pulses containing the temporal information provides selection of gamma-ray pairs in cascade, while selection on the basis of energy is effected through analysis of slow pulses. The two fast pulses appear at discriminators that in turn produce standardized pulses fed to a time-to-pulse-height converter. If the two standardized pulses appear at this converter within 200 ns of

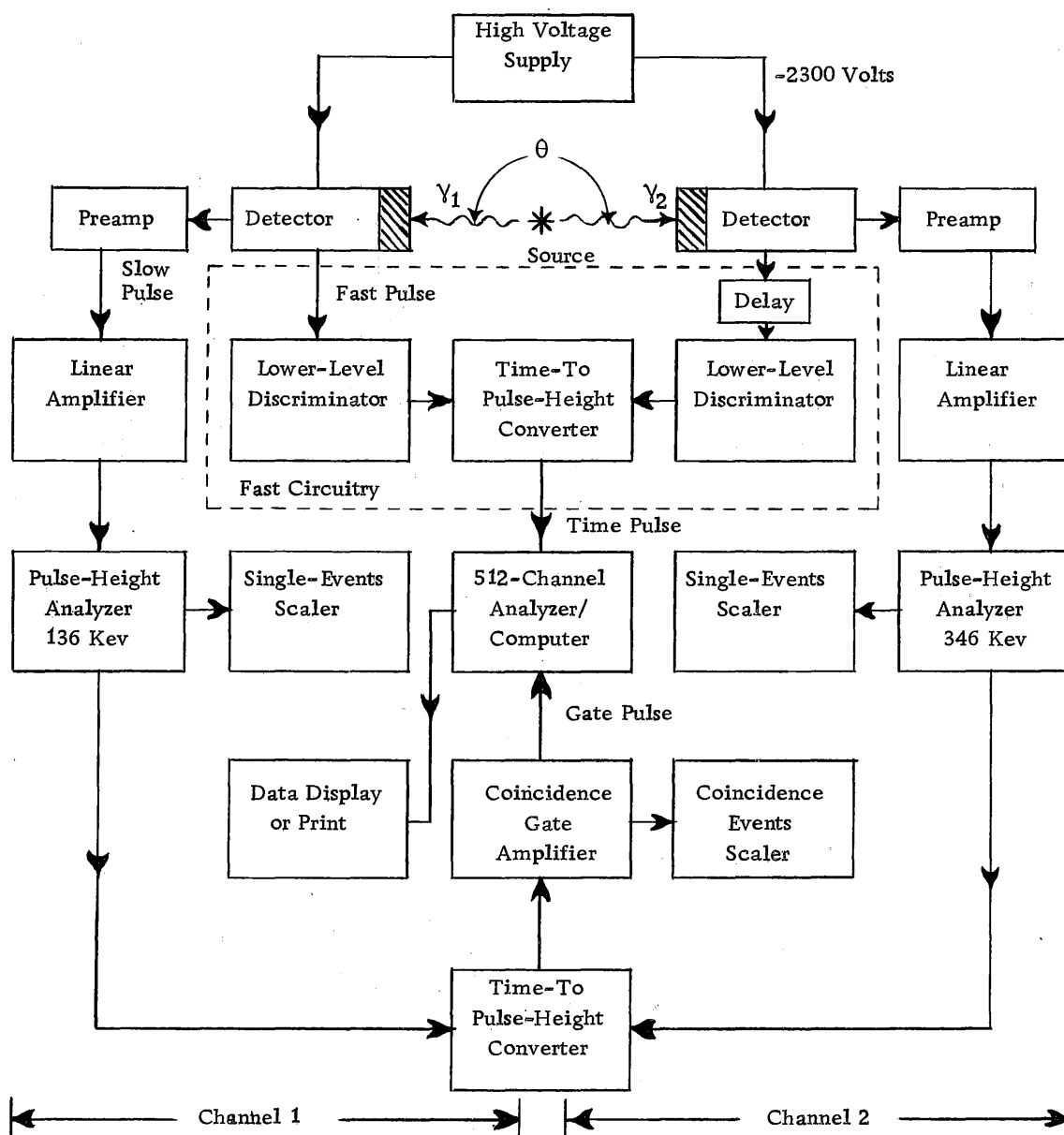


Figure 3. Block diagram of the apparatus. Circuitry within the dotted lines processes fast-rise pulses carrying the time information.

each other, a pulse is produced whose amplitude is directly proportional to the time interval between the emission of γ_1 and γ_2 . This pulse is presented to a gating circuit in a 512 channel pulse-height analyzer to await processing of the slow pulses. The two slow pulses, whose amplitudes are directly proportional to the energies of γ_1 and γ_2 are fed through linear amplifiers to single channel pulse-height analyzers set to accept only pulses corresponding to gamma-ray energies of 346 Kev and 136 Kev. When these analyzers receive acceptable pulses they produce standardized pulses fed to a second time-to-pulse-height converter. If a pulse from channel 2 arrives at this converter within 800 ns of a similar pulse from channel 1, a coincidence gate pulse is produced indicating that both γ_1 and γ_2 of correct energy have been detected. This gate pulse, shaped by an amplifier, opens the gating circuit of the 512 channel analyzer to allow the pulse from the fast circuitry containing the time information to be analyzed and stored. The 512 channel analyzer stores this time-analog voltage pulse according to its amplitude in one of 512 channels representing equal voltage increments. A calibration relating these voltage increments to the time intervals between the emission of gamma rays γ_1 and γ_2 then allows the temporal data to be recovered in the form of a delayed coincidence spectrum. The angle between the propagation vectors of detected gamma rays is obtained by means of a protractor

mounted on the spectrometer table upon which the source, fixed and movable detectors are mounted. The angles can be determined to within 0.25 degree.

Test and Calibration

Operational checks of the apparatus included the measurement of time resolution, time calibration and linearity for the conditions under which the experiment was performed. These measurements were obtained by observing 512 Kev gamma-ray pairs in prompt coincidence produced by positron annihilation in Na^{22} . The time resolution T of the system was 2.6 ns as defined by the half width at half maximum of the prompt coincidence curve shown in Figure 4. This data was obtained with the pulse height analyzers set to accept events produced by gamma rays losing energies of ~ 346 Kev and ~ 136 Kev due to Compton scattering in the detectors. Linearity and time calibration information was obtained by delaying the fast pulse of channel 2 by 5.0 ns increments and recording a prompt curve for each increment. Stability of the apparatus during the experiment was assessed by noting excursions in the peak channel number of the coincidence data and by recording the pulse height analyzer settings determined periodically. Long-term drift was minimized by regulation of line voltage and ambient temperature and by thorough warm-up followed by continuous operation for the duration of the experiment.

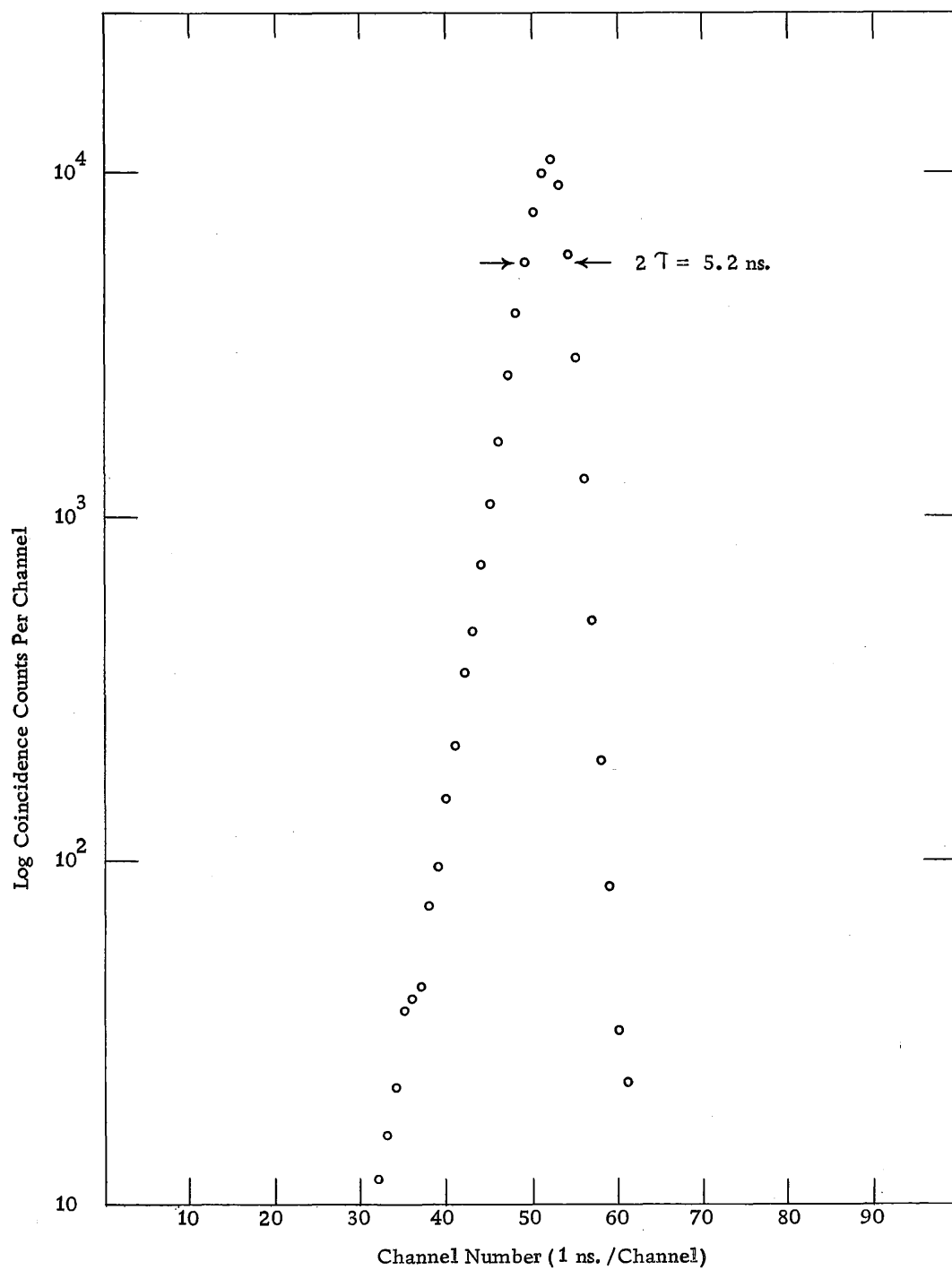


Figure 4. Prompt coincidence curve showing time resolution of the apparatus.

III. PROCEDURE

The measurement of the directional correlation of the 346 Kev-136 Kev cascade entails adjustment of the apparatus so as to accept events of these energies. The desired settings of the pulse-height analyzers for this condition are indicated by the energy acceptance windows ΔE shown in Figure 5. All gamma rays that transfer to the detectors energies falling within these energy windows can contribute to the collection of coincidence data making interpretation of these data difficult. When channel 1 is set to accept 136 Kev gamma rays, it will also accept 133 Kev gamma rays since the apparatus cannot resolve these energies. Furthermore, when channel 2 is set to accept 346 Kev gamma rays, it will respond to those 482 Kev gamma rays that lose energies of ~ 346 Kev due to Compton scattering in the detector as well. Because each cascade satisfies the coincidence requirement according to the decay scheme of Figure 1, the two competing cascades 133-482 Kev, 133-346 Kev as well as the 346-136 Kev cascade of interest contribute to the composite coincidence data shown in Figure 6. Although in the case of the simple cascade discussed in the theory only the slow circuitry is needed to obtain $W(\theta)$, the fast circuitry is essential to this experiment because of the presence of these competing cascades. That is, the fast circuitry provides just the information needed to extract the

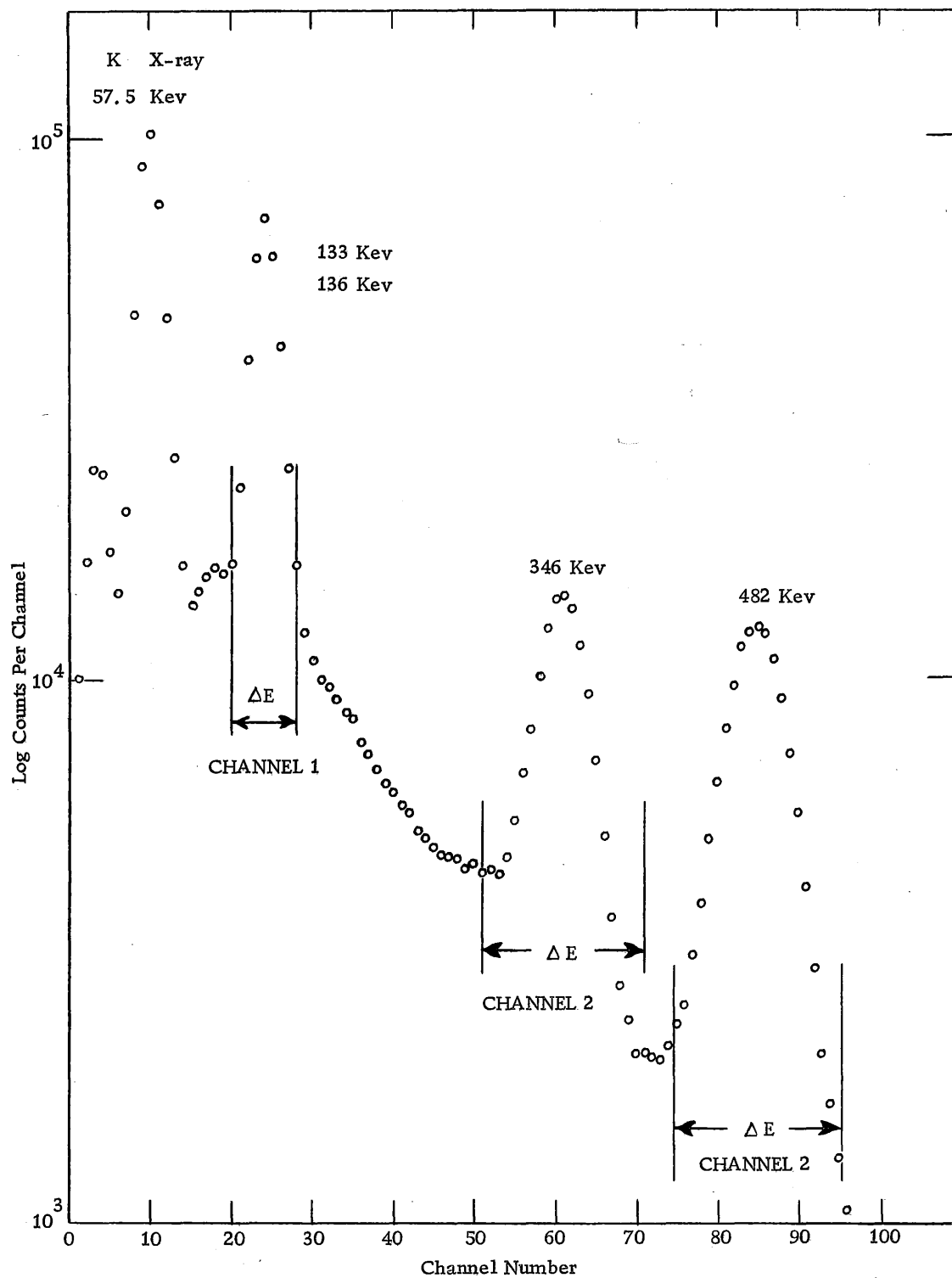


Figure 5. Partial gamma-ray energy spectrum of Ta^{181} showing energy windows ΔE .

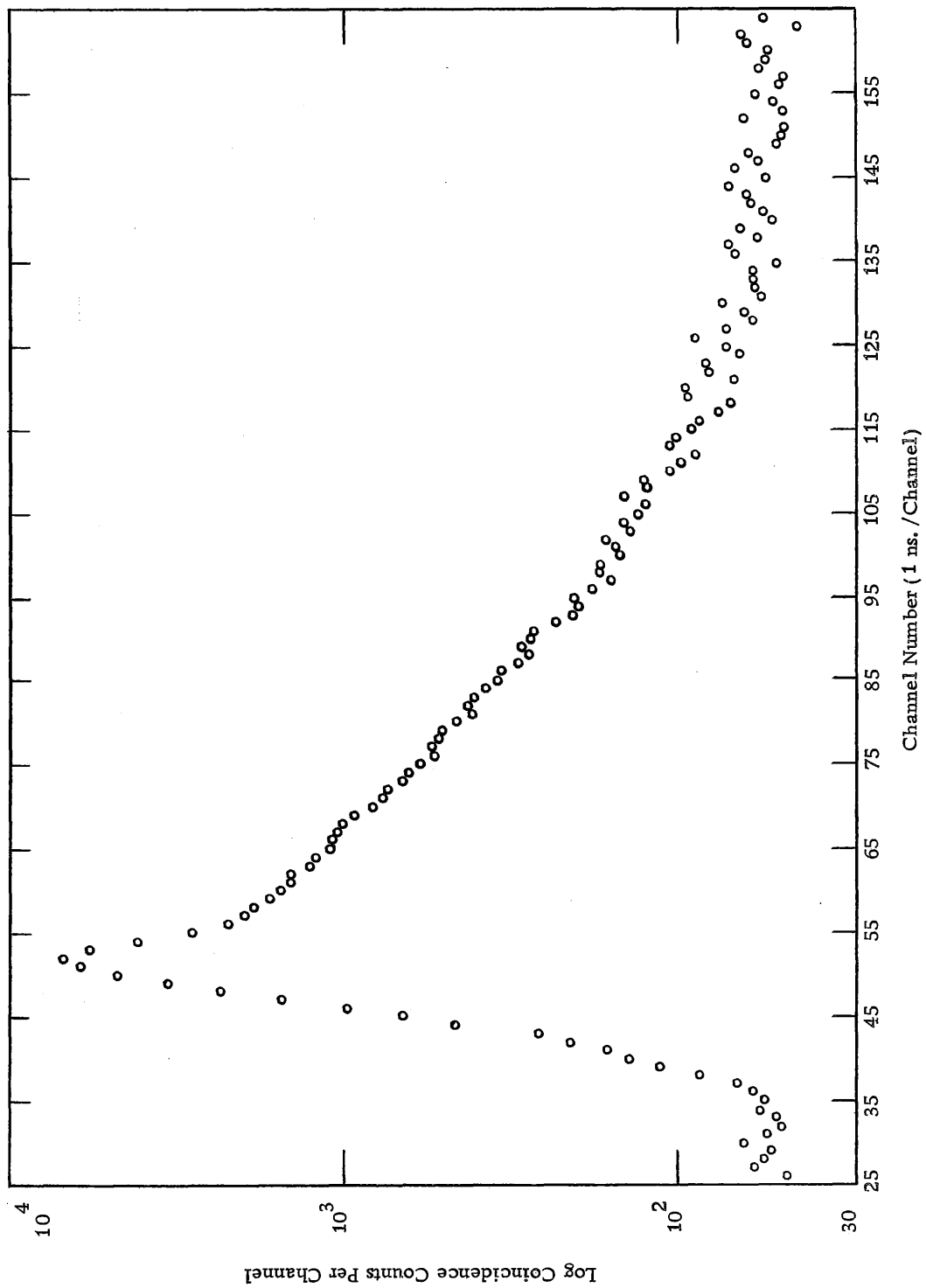


Figure 6. 133-346 Kev composite coincidence data containing both delayed and prompt components.

346-136 Kev coincidence events from the composite data.

The 346-136 Kev coincidence events were recovered from the composite data in the following way. With the pulse-height analyzer windows set on 133 Kev and 346 Kev as previously stated, composite coincidence data were collected over a two-hour period at a given angle θ and stored in one half of the memory of the 512-channel analyzer. Channel 2 was then set to accept 482 Kev gamma rays with the result that only 133-482 Kev pure delayed coincidence data were collected and stored in the other half of the memory. Figures 6 and 7 illustrate these data for a typical run. By means of the computer function of the 512-channel analyzer, an appropriate amount of the pure 133-482 Kev data of Figure 7 was subtracted channel-by-channel from the composite data of Figure 6 to obtain the 346-136 Kev component illustrated in Figure 8. Subtraction of the correct amount of 133-482 Kev data was indicated when the count in channels containing only the delayed coincidence data from the competing cascades fell to the background level.

The validity of this procedure rests on the assumption that the delayed coincidence spectrum of the 133-482 Kev cascade has the same time distribution as the two interfering delayed coincidence components in the composite data.

The data acquisition program during the experiment consisted of observations at angles θ in 10 degree increments from 90 degrees

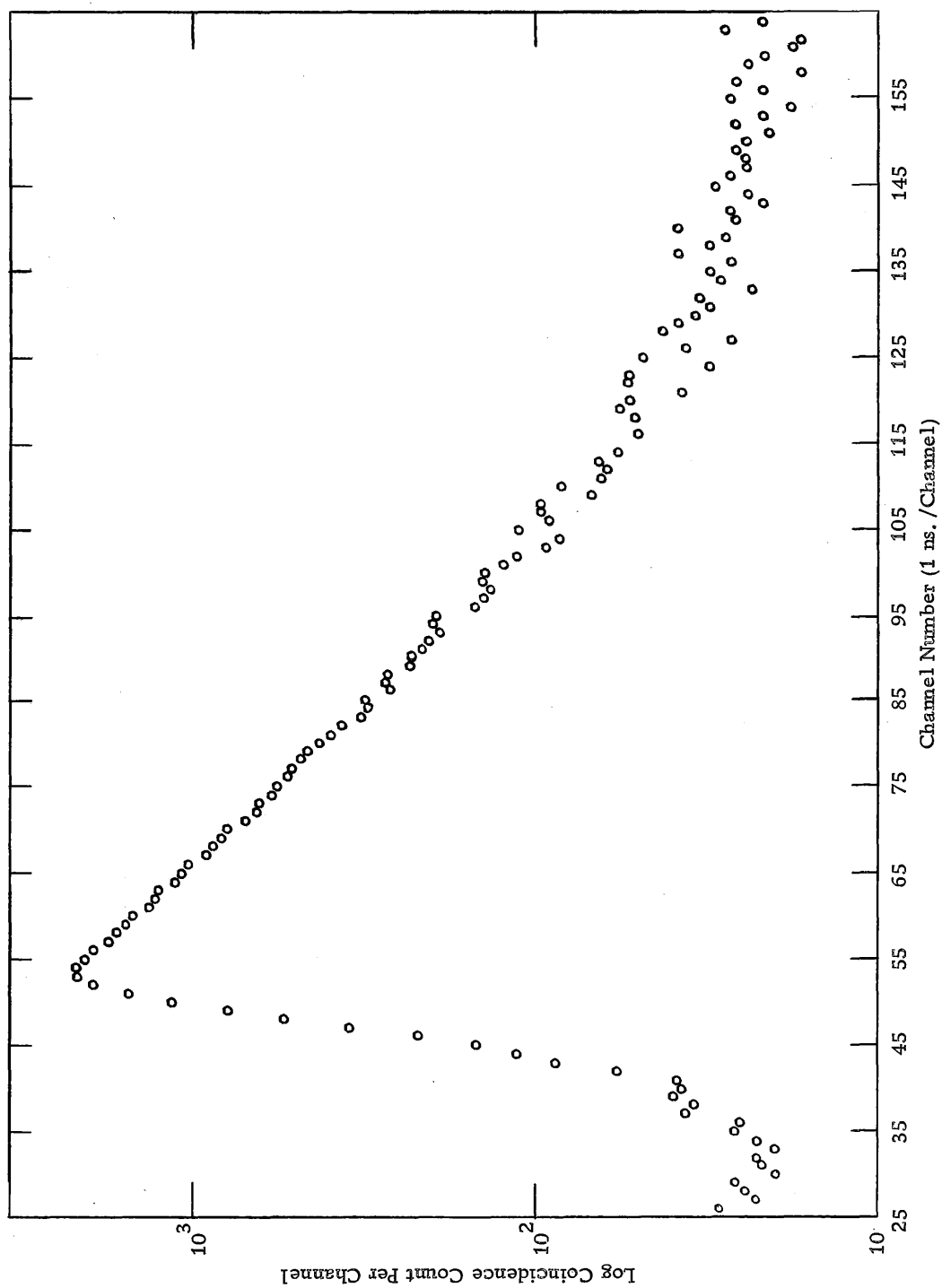


Figure 7. The 133-482 Kev pure delayed coincidence data.

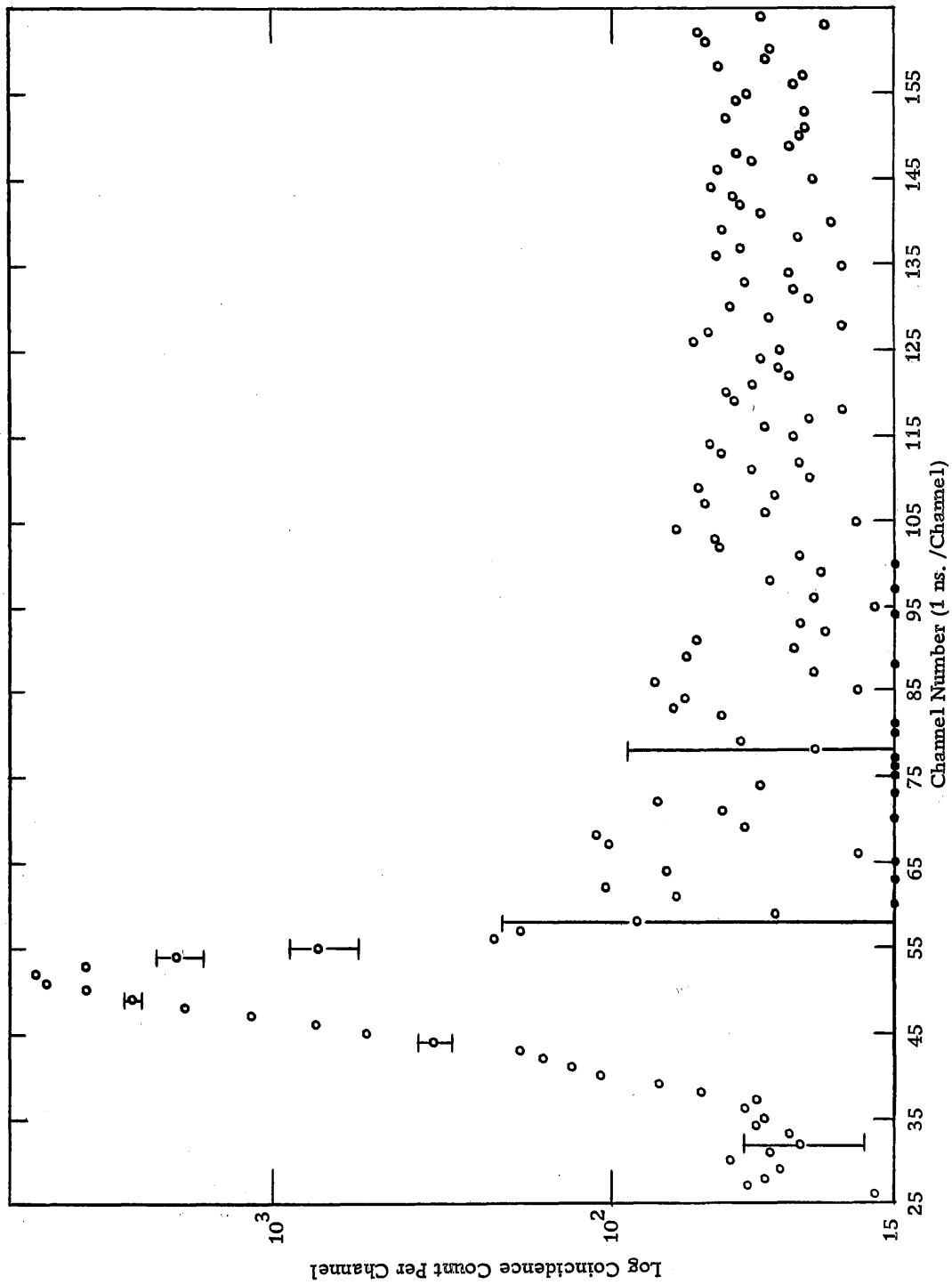


Figure 8. The 346-136 Kev component. Filled circles indicate counts less than 15.

to 270 degrees using the sequence 180° , θ , 180° , $-\theta$. For each observation composite data were taken, 133-482 Kev data were taken and the subtraction was made. Scaler counts were recorded and data printed out. Four sequential observations for each of nine angles $|\theta|$ over a two week period provided the data.

There was yet another source of coincidence events to be considered as contributing to the 346-136 Kev component. A calculation indicated that a 482 Kev gamma ray could Compton-scatter out of the detector of channel 2 losing an energy of ~ 346 Kev, and carry the remainder of its energy, ~ 136 Kev, into the detector of channel 1. This possibility necessitated the performing of a subsidiary experiment in which prompt coincidence data were collected using a Be^7 source that produced a single gamma ray of 478 Kev. The pulse-height windows were set on 133 Kev and 346 Kev and the resulting data provided a non-isotropic back-scatter correction factor to be applied to the 346-136 Kev data.

DATA REDUCTION

A numerical integration over 26 channels centered on the peak channel of the 346-136 Kev coincidence data obtained for each angle θ provided the total coincidence count. The contribution of accidental coincidences to the total count was found by an integration over the background count in channels far removed in time from the peak channel. This chance coincidence component and the appropriate component for the back-scatter correction were subtracted from the total count to yield the true coincidence count. These chance coincidence and back-scatter corrections involved subtractions of 3 to 4 percent and about 17 percent respectively from the total count. The true coincidence count was normalized to the product of counts obtained by the two singles scalers during collection of the parent composite data. Ratios were formed by dividing the normalized true coincidence count for each angle θ by the count obtained from the preceding 180 degree observation, and these ratios were averaged to obtain a ratio for each of nine angles $|\theta|$. The experimental coefficients A_{22} and A_{44} were obtained from a weighted least-squares fit of the theoretical function $W(\theta)/W(\pi)$ to the nine ratios using weighting factors based on their statistical errors. These coefficients

were corrected for finite angular resolution of the detectors through the application of appropriate correction factors compiled by Yates (5, p. 1695) and were found to be

$$A_{22} = 0.190 \pm 0.011, \quad A_{44} = -0.025 \pm 0.024.$$

Figure 9 displays the agreement between the measured values of $W(\theta)$ and the curve generated by the theoretical function $W(\theta)$ using the experimentally determined coefficients A_{22} and A_{44} .

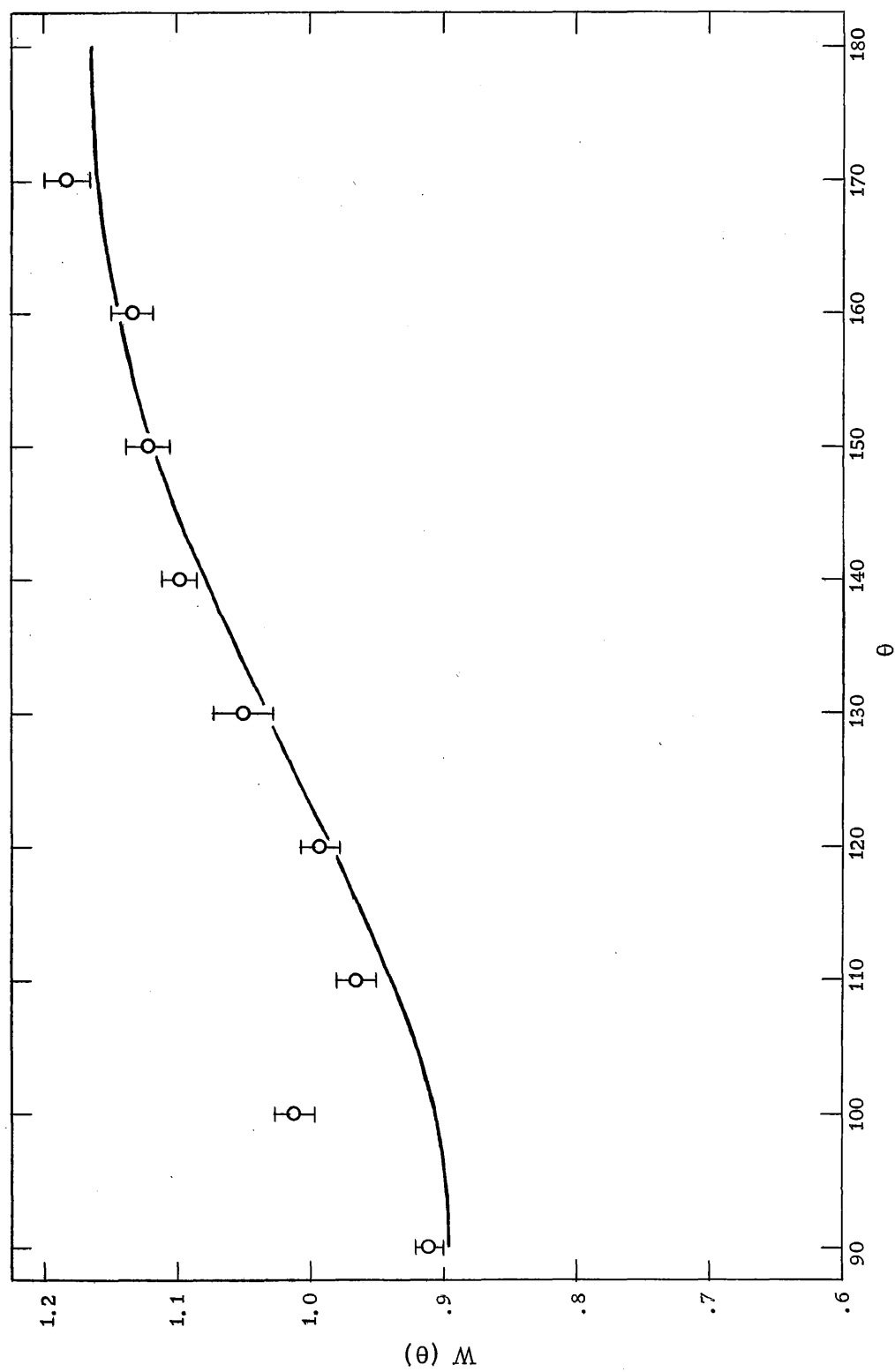


Figure 9. Least-squares fit of $W(\theta)$ to the data.

CONCLUSION

The directional correlation of the 346-136 Kev gamma-gamma cascade in Ta¹⁸¹ has been measured using the technique of fast delayed coincidence scintillation spectrometry. The correlation coefficients A_{22} and A_{44} thus obtained are compared in Table I with those obtained from directional correlation measurements performed by McGowan and by Debrunner et al. and with those calculated on the basis of data obtained from conversion electron measurements carried out by other investigators (9-13; 1, p. 6-6-142).

TABLE I

DIRECTIONAL CORRELATION COEFFICIENTS AND PERCENTAGE
ADMIXTURES FOR THE 346-136 Kev CASCADE IN Ta¹⁸¹

Reference	A_{22}	A_{44}	Percent E2	
			from A_{22}	from A_{44}
McGowan (2)	0.202±0.009	-0.053±0.014	17.6±1.1	33.7± 8.9
Debrunner (3)	0.17 ±0.04	0.01 ±0.04	14.1±1.9	6.4±25.4
Current work	0.190±0.011	-0.025±0.024	16.2±1.2	16 ±15
From conversion electron measurements	.193	-0.0264		17

The spin assignments and multipole orders shown in the decay scheme of Figure 1 have been confirmed by other methods (1, 6-6-140-145) since Debrunner's work and are firmly established. Taking the spin sequence $5/2(E2)9/2(M1 + E2)7/2$ to be correct and following Arns and Wiedenbeck (14), the percentage admixture of E2 radiation in the second gamma transition of the 346-136 Kev cascade was determined with the aid of the parametric plot shown in Figure 10. The mixed transition was found to contain $16.2\% \pm 1.2\%$ E2, a value that is in excellent agreement with the 17% E2 obtained from conversion electron measurements. For purposes of comparison, the E2 admixtures implied by the correlation coefficients of McGowan and Debrunner were calculated by the same method used in the current work and are included in Table I. As indicated in Figure 10, both coefficients obtained in the present experiment are consistent with the 16.2% E2 admixture in the 136 Kev transition. Although Debrunner's results are also consistent in this sense, the large uncertainties prohibit a very precise assignment of the E2 admixture. The coefficients of McGowan however are clearly inconsistent with the established spin assignments since the E2 admixtures obtained on the basis of these assignments from his A_{22} and A_{44} do not agree.

In recapitulation, the results of this experiment are consistent

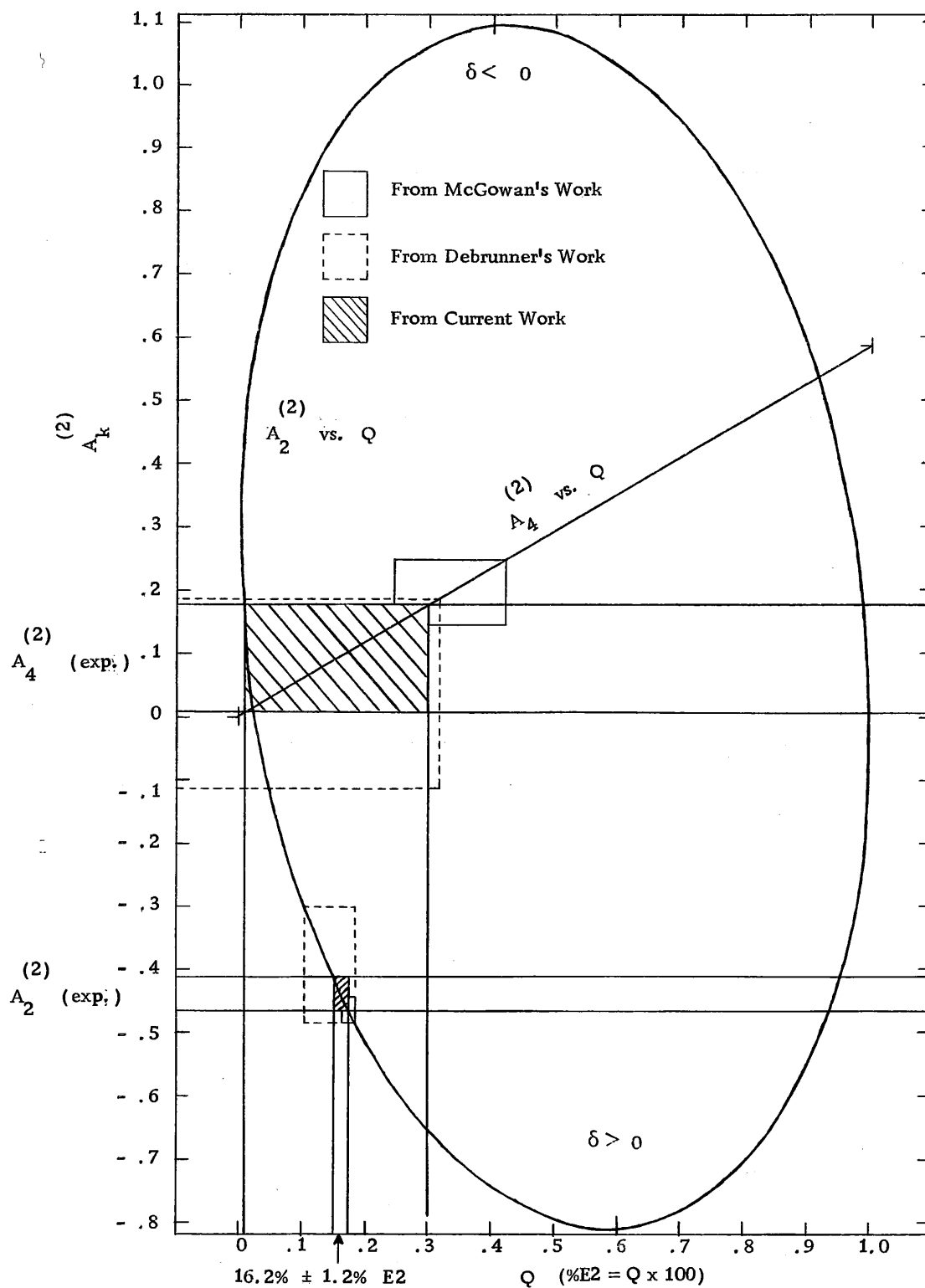


Figure 10. E2 admixture in the 136 Kev transition implied by the A_{22} and A_{44} of McGowan, Debrunner et al. and the current work.

with the spin assignments, multipole orders and mixing ratio of the 346-136 Kev cascade as established. In addition, the ambiguity in the mixing ratios from previous measurements has been resolved. Finally, the successful performance of this experiment has provided a basis of operational technique and analytical procedure with which similar experiments can be confidently undertaken.

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