# Measurements of Short-Lived Isomers*

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The half-lives of eight short-lived isomers were investigated by the delayed coincidence method of detecting short-lived excited gamma-states following beta-emission. Of these, the half-life of $Hg^{198*}$ following beta-decay of $Au^{198}$ was measured as $2.3\times 10^{-8}$ sec. Excited states of $Mg^{24*}$, $Hg^{199*}$, $Ca^{42*}$, $Nd^{142*}$, $A^{38*}$, $Te^{122*}$, and $Fe^{56*}$ were found to have half-lives shorter than 1 to $2\times 10^{-8}$ sec., the lowest value measurable, owing to circuit limitations.

By means of the measured half-lives or upper limits, probable $l$ values, the change in units of angular momentum, were assigned through comparison with the theoretical values of the Segrè-Helmholz formula. Correlation with $l$ values estimated through other methods has resulted in probable assignment of $l$ values for four isomers and upper $l$ values for the remaining four.

## I. INTRODUCTION

THE measurement of short-lived nuclear isomers by the method of delayed coincidences has been recently employed by many investigators.[1–3] With the development of scintillation counters of fast decay time, half-lives of the order of $10^{-8}$ to $10^{-9}$ second can be measured.[4–6]

The delayed coincidence method refers to the measurement of the time interval between the formation of the excited state and emission of the gamma-ray from the decay of the state. In the following work the time reference for the formation of the state is taken to be the emission of a beta-particle leading to the excited state. The general relationship for the number of coincidences per second as a function of delay time, $T$, as the pulse from the beta-detector is delayed is thus:

$$C=2\tau N_1 N_2 \lambda e^{-\lambda T},$$

where $\tau$ is the resolving time of the coincidence circuit, $\lambda$ is the decay constant, $N_1$ is the counting rate of the beta-particles, and $N_2$ is the number of gamma-rays per beta-disintegration. This equation neglects the variation of pulses in the detector or assumes that this variation is very small with respect to the delay time.

Van Name[7] has shown the value of the consideration of time of pulse formation in the interpretation of results from delayed coincidence counting. By assuming a triangular distribution of pulses about the mean time of pulse formation, Van Name derived directly the variation of coincidences as a function of delay time. Binder[8] has developed an analysis similar to Van Name's by assuming a Gaussian spread for the pulses. Computations from this assumption are simple, since one expression is valid for the entire range of the curve.

In all three expressions the coincidence rate approximates an exponential function for large delay times, which relationship has also been shown by Newton.[9] The analyses show that a plot of the logarithm of the counting rate *versus* delay time will fall off with a slope of $\lambda$, this method being used for calculation of half-life in this work. The entire curve has been fitted by Binder[8] and the upper limit estimated from the constants of the theoretical curve with the shortest half-life that deviates appreciably from the symmetric curve. This limit has been placed at 1 to $2\times 10^{-8}$ second, dependent on the resolution. As a measured half-life of $2.3\times 10^{-8}$ sec. deviates markedly from the symmetric curve, the writer feels this limit is justified experimentally.

## II. CORRELATION WITH THEORETICAL PREDICTION OF ISOMERIC HALF-LIFE

A simple relationship between the decay constant, $\lambda$, and $l$, the change in angular momentum, has been previously derived by Segrè and Helmholz.[10,11] Segrè and Helmholz point out that their assumption that the dimension of the radiation multipole be equal to the nuclear radius ($A^{\frac{1}{3}}\times 1.45\times 10^{-13}$ cm) is only a crude approximation; but since a change of angular momentum of one unit in $l$ may result in a difference of predicted half-life by a factor of $10^5$, this formula is sufficient to assign $l$ values to the gamma-ray transition. With so great a change, a difference in the measured value by as much as a factor of 100 would not prohibit the $l$ assignment. Such assignment is not necessarily definite, but merely the closest fit to the theoretical prediction. This formula in general predicts somewhat too large a value for the half-life; therefore, in half-

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1 S. DeBenedetti and F. K. McGowan, Phys. Rev. **74**, 728 (1948).
2 Bowe, Goldhaber, Hill, Meyerhof, and Sala, Phys. Rev. **73**, 1219 (1948).
3 P. T. Bittencourt and M. Goldhaber, Phys. Rev. **70**, 780 (1946).
4 F. K. McGowan, Phys. Rev. **77**, 138 (1950).
5 M. Deutsch and W. G. Wright, Phys. Rev. **77**, 139 (1950).
6 R. E. Bell and R. L. Graham, Phys. Rev. **78**, 490 (1950).
7 F. W. Van Name, Jr., Phys. Rev. **75**, 100 (1949).
8 D. Binder, Phys. Rev. **76**, 856 (1949).
9 T. D. Newton, Phys. Rev. **78**, 490 (1950).
10 A. C. Helmholz, Phys. Rev. **58**, 48 (1941).
11 E. Segrè and A. C. Helmholz, Rev. Mod. Phys. **21**, 271 (1949).

lives falling between two $l$ values, the shorter predicted half-life is preferred.

## III. DESCRIPTION OF EQUIPMENT

For the detection of beta-particles, cylindrical proportional counters were used. The length measured five-eighths inch, with a one-quarter-inch inside diameter centered about a three-mil coaxial wire. The small size resulted in a low geometrical efficiency, but was designed to keep the random fluctuations at a minimum. These counters were used by Farley[12] in detection of heavy particles (protons, deuterons, tritons, and alpha-particles). By resolution curves, Farley[12] has shown the pulses to be very sharp, rising at least as fast as $5\times10^{-8}$ sec.

For the detection of gamma-rays a scintillation counter with selected 931A electron multiplier tube was used, operating at dry ice temperature. Although naphthalene and calcium fluoride crystals were initially tried as scintillation phosphors, both exhibited decay times which were too long[13] for use. Polystyrene showed the fastest resolving time and a decay time too fast to be detected with this equipment. The pulses from polystyrene were extremely small and hard to detect,[14] but at the time of these measurements the faster organic crystals such as stilbene were not generally available.

As a preamplifier for the electron multiplier tube the cathode follower shown in Fig. 1 was used. This is similar to the cathode preamplifier used by Farley except for the resistance network used to apply voltage steps to the 931A electron multiplier tube. This circuit has a wide band width (over 25 Mc/sec.); and although the gain is low, this is no disadvantage since greater amplification is available in the linear amplifier than is required.

Although a similar cathode follower was originally used with the proportional counter for heavy particle

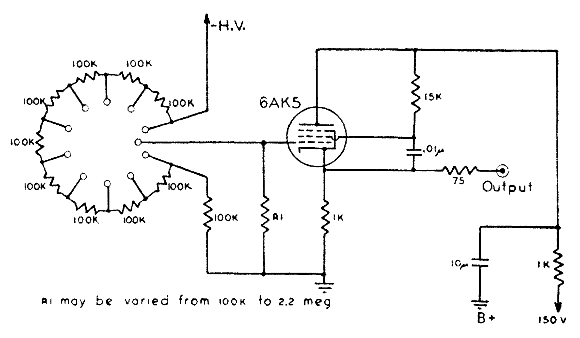


FIG. 1. Electron multiplier preamplifier for scintillation counter. Outputs of both preamplifiers are carried through RG11U coaxial cable to amplifiers of 75-ohm input.

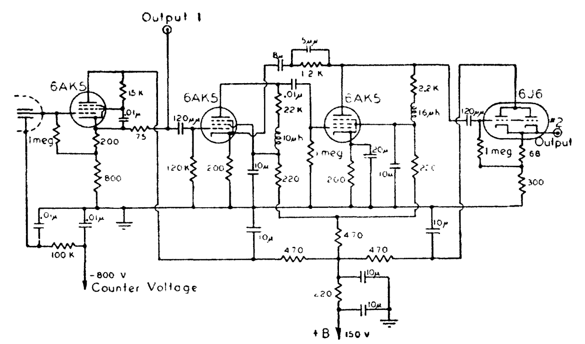


FIG. 2. Preamplifier for proportional counter. Output #1 is used for detection of heavy particles. Output #2 includes one stage of amplification and is used for detection of $\beta$-particles.

detection, the smaller pulses from beta-particles made the addition of one section of amplification necessary. After the cathode follower for impedance matching, one section of the wide-band amplifier developed by Schultz[15] was added. The band width of these pairs is 23 to 26 Mc/sec. and, as the output is again a cathode follower for line matching, the entire preamplifier is of this band width. This circuit is shown in Fig. 2. The linear amplifiers themselves consist of sections of these feed-back pairs terminated by the 6J6 cathode-follower output of Fig. 2.

The coincidence circuit was developed by Farley and possesses a threshold delay time of the discriminator of approximately $2\times10^{-9}$ sec. With the counters described, resolution as low as $10^{-8}$ sec. has been reached; but $3.8\times10^{-8}$ sec. is usual.

For a variable delay, lengths of seventy-ohm coaxial cable (RG 11/U) was used. The velocity of a pulse in this line has been found to be $2\times10^{10}$ cm/sec.; thus, a convenient length of two meters of this cable represents a time delay of $10^{-8}$ sec.

## IV. RESULTS OF ISOMER MEASUREMENTS

### A. $Na^{24}$

While early investigation on the disintegration of $Na^{24}$ gave rise to conflicting reports, most of the later work is in general agreement with Siegbahn,[16] indicating a beta-ray group of 1.39-Mev peak energy and two gamma-rays of 1.38 and 2.76 Mev.

The $Na^{24}$ source was placed between the proportional counter and the polystyrene counter with a brass absorber screening the gamma-scintillation counter from the beta-particles. Since the proportional counter has a low efficiency for gamma-rays, this arrangement resulted in the detection of beta-particles alone by the proportional counter and gamma-rays alone by the scintillation counter. This detection is important for this case, which involves cascaded gamma-rays, since it proves the data obtained are for $\beta-\gamma$ coincidences and not $\gamma-\gamma$.

[12] B. G. Farley (to be published). Ph.D. thesis, Yale University, 1948.

[13] W. J. MacIntyre, Phys. Rev. **75**, 1439 (1949).

[14] G. N. Harding of the Atomic Energy Research Establishment at Harwell, England, has estimated that the pulses from polystyrene were about one-tenth the size of those from naphthalene (private communication).

[15] H. L. Schultz, "High speed counter and short pulse techniques." Brookhaven Conference Report, p. 35, (August 1947).

[16] K. Siegbahn, Phys. Rev. **70**, 127 (1947).

The resulting curve from the measurement of coincidence rate *versus* delay time is shown in Fig. 3. The coincidence rate for this measurement was of the order of 50 to 200 coincidences/min. at the peak, with background (accidental coincidences) at 8 to 40 coincidences/min. The accidental coincidence rate on all this work has been measured directly by putting a large delay in the gamma-ray channel so as to get far beyond the resolution curve of true coincidences. This method is believed to be superior to the calculation of accidental coincidence rate, $A$, by the relation of $A=2\tau N_1N_2$, where $\tau$ is the resolving time, and $N_1$ and $N_2$ are the single rates in both channels. This formula makes assumptions both on the circuit and the invariance of $\tau$.

Although the curve of Fig. 3 shows a slight asymmetry to the trailing edge, it is too fast for this equipment to measure, and gives essentially a resolution curve as obtained by simultaneous stimulation of both counters. Measured states as fast as $2\times10^{-8}$ sec. show a marked asymmetry; in addition, the theoretical work of Binder has shown that states with a half-life as low as $1\times10^{-8}$ sec. can be detected by the change in shape of the coincidence rate curve. From this observation it is estimated that the excited states of $Mg^{24*}$, formed by beta-decay of $Na^{24}$, must have a half-life shorter than $1\times10^{-8}$ sec.

The zero point for delay time is arbitrary, since the amplifiers and other circuits are not balanced exactly. Also, the placement and shape of the source may cause shifting of the zero point, which may explain the variation of the zero point in the succeeding resolution curves.

For the gamma-ray of energy 1.38 Mev, the Segrè-Helmholz formula predicts a half-life of $6\times10^{-13}$ sec. for $l=2$; $2\times10^{-8}$ sec. for $l=3$; and $1\times10^{-4}$ sec. for $l=4$, indicating an $l$ change of three or less, with three itself doubtful.

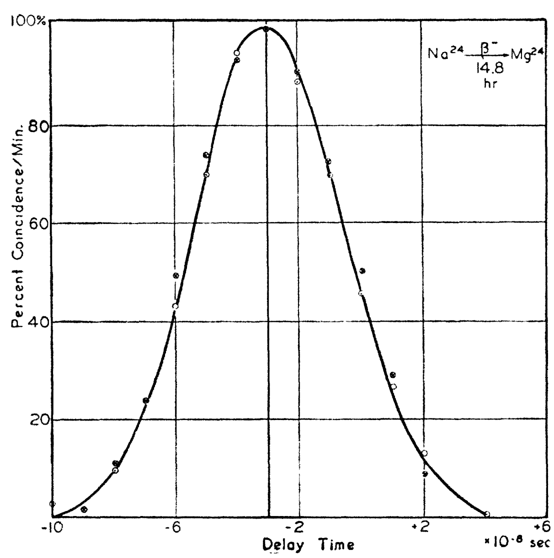


FIG. 3. Coincidence rate *versus* delay time curve for $Na^{24}$ decay. Plus values of delay time refer to delay added to $\beta$-channel. Symmetry of the curve indicates the state to be too fast to be measured by this equipment.

From the lack of sharp resonance absorption in scattering experiments on these gamma-rays, Pollard and Alburger[17] have shown that the radiation must involve an $l$ value of two or higher. From a combination of these two experiments, it seems probable that the transition of both gamma-rays is $l=2$. This result is also consistent with the conclusions of Brady and Deutsch,[18] who have measured the angular correlations of the gamma-rays and placed the spin of the levels in $Mg^{24*}$ at 4, 2, and 0, a difference of two between the levels.

## B. $Cl^{38}$

Although the disintegration scheme of $Cl^{38}$ is complex, most measurements have been consistent with the scheme of Hole and Siegbahn,[19] except for small variations in energies and relative intensities. This work shows a direct beta-transition, a second beta-group followed by one gamma-ray, and a third beta-group followed by one gamma-ray transition to the second gamma-ray transition in cascade.

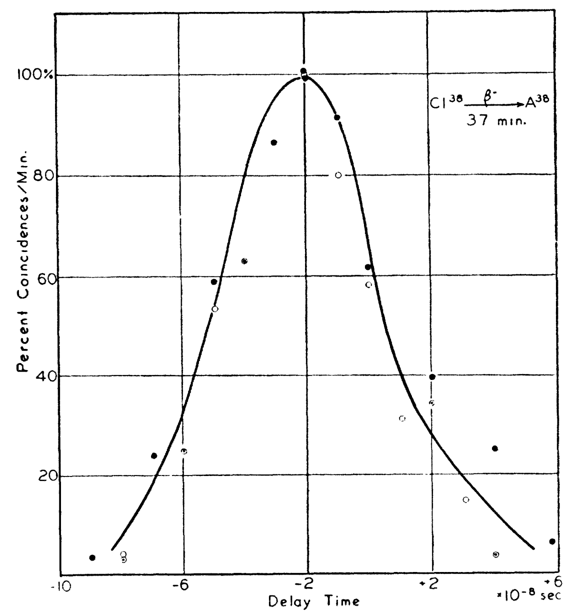


FIG. 4. Coincidence rate *versus* delay time curve for $Cl^{38}$ decay. The point of zero delay time is arbitrary, since the two channels are not necessarily balanced.

The two gamma-transitions have comparable intensities and energies, and thus would contribute approximately equal effects on the curve. In the event a measurable half-life had been found, additional absorption experiments would have been necessary in order to identify to which transition this half-life belonged. Figure 4 is seen to be symmetric, however, which shows

[17] E. C. Pollard and D. E. Alburger, Phys. Rev. 74, 926 (1948).
[18] E. L. Brady and M. Deutsch, Phys. Rev. 74, 1541 (1948).
[19] N. Hole and K. Siegbahn, Arkiv. Mat. Astron. Fys. 33A, No. 9 (1946).

the transition half-lives to be less than $1\times10^{-8}$ sec.

For the 1.60-Mev gamma-ray of $A^{38*}$, the Segrè-Helmholz equation shows a half-life of $2\times10^{-13}$ sec. for $l=2$; $2\times10^{-9}$ sec. for $l=3$; and $1\times10^{-5}$ sec. for $l=4$. Similarly, for the 2.12-Mev gamma-ray a half-life of $4\times10^{-10}$ sec. for $l=3$ and $6\times10^{-7}$ sec. for $l=4$ is predicted. From this prediction the transitions are assigned an $l$ value of equal to or less than three.

Hole and Siegbahn[19] have assigned spin values for the various levels of $A^{38}$; and, from the degree of forbiddeness of the beta-particle transitions from $Cl^{38}$ decay, have concluded that both the ground level and the second excited level have spins of zero and the first excited level a spin of one or two. This conclusion leads to an $l$ value equal to or less than two for both gamma-

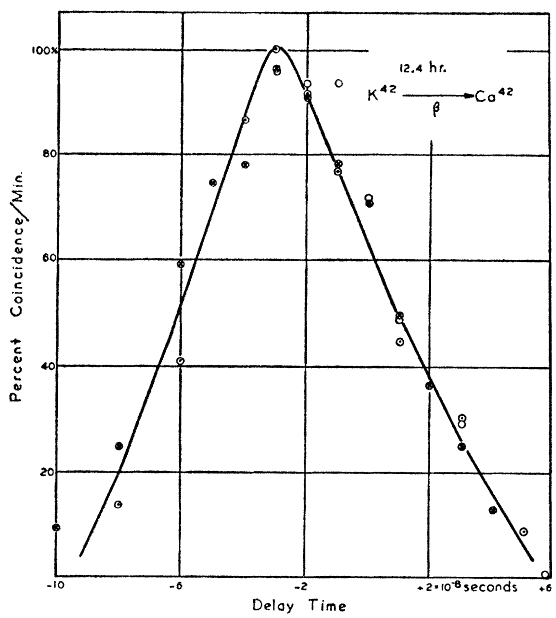


FIG. 5. Coincidence rate *versus* delay time curve for $K^{42}$ decay.

transitions, this value being consistent with the above measurement.

Langer's[20] interpretation from his measurements places the spin of the ground level of $A^{38}$ at zero and the second excited level of $A^{38*}$ at three. This scheme is also consistent with the values obtained from the upper half-life limit.

The curve of Fig. 4 is seen to be less clearly defined than that of Fig. 3. This difference is probably the effect of poorer statistical accuracy, since the short half-life of $Cl^{38}$ necessitated coincidences taken per number of single counts at low counting rates where accidentals were negligible. As the accidentals vary as the second power of the counting rate, it is advantageous to minimize their effect so that decay of the parent state may be easily accounted for.

[20] L. M. Langer, Phys. Rev. **77**, 52 (1950).

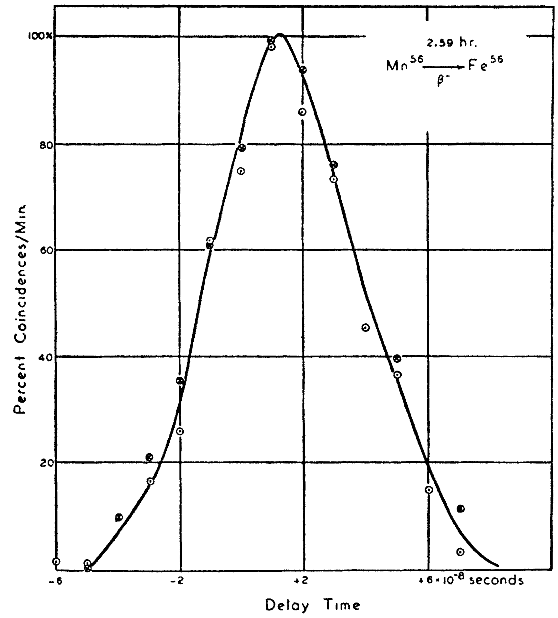


FIG. 6. Coincidence rate *versus* delay time curve for $Mn^{56}$ decay.

## C. $K^{42}$

The decay of $K^{42}$ involves only one gamma-ray of 25 percent intensity as given by Siegbahn.[21] Good counting statistics were hard to obtain, since the strong (75 percent) beta-particle transition to ground state raised the background of accidental coincidences.

The data are plotted in Fig. 5 and show a predominately symmetric curve. The resolution of this curve is not as sharp as the previous curves (probably owing to the strong beta-transition), so that the upper limit of the half-life is placed at less than $2\times10^{-8}$ sec. rather than less than $1\times10^{-8}$ sec., to which the sharper resolution curves can be read.

Theoretical predictions from the Segrè-Helmholz equation show a half-life of $2\times10^{-13}$ sec. for a value of $l=2$; $3\times10^{-9}$ sec. for $l=3$; and $1\times10^{-5}$ sec. for $l=4$. This prediction indicates a value for $l$ of possibly three but more probably two or less.

In an interpretation of $K^{42}$ radioactivity by Shull and Feenberg[22] from an analysis of Siegbahn's data for a characteristic forbidden type of energy distribution of the beta-transition, the value of $l$ for the transition of $Ca^{42*}$ was placed at two. The combination of investigations show that $l$ is probably two.

## D. $Mn^{56}$

In spite of the complexity of the transitions and the varied types of investigations, almost all data obtained on $Mn^{56}$ decay are consistent with the decay scheme advanced by Elliot and Deutsch,[23] and by Siegbahn

[21] K. Siegbahn, Arkiv. Mat. Astron. Fys. **34B**, No. 4 (1948).
[22] F. B. Shull and E. Feenberg, Phys. Rev. **75**, 1768 (1949).
[23] L. G. Elliot and M. Deutsch, Phys. Rev. **64**, 321 (1943).

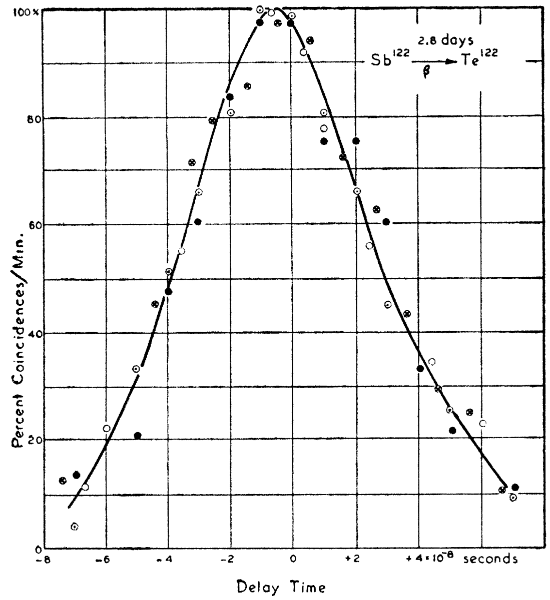


FIG. 7. Coincidence rate *versus* delay time curve for Sb$^{122}$ decay.

and Johannson.[24] This scheme shows all excited levels decaying to the ground state by the path of the 0.822-Mev gamma-transition. The ratio of the intensity of this transition to that of the next strongest transition is then 10:3, so that the measurement will be dominated by this 0.822-Mev gamma-ray.

Good counting statistics were obtained on this measurement with a rate of 1000 to 1500 coincidences/min. This rate is shown by the well-defined curve of Fig. 6, which is also seen to be symmetric and indicates a half-life less than $1\times10^{-8}$ sec. Comparison with the Segrè-Helmholz formula yields a prediction for an $l$ value of two or less.

Elliot and Deutsch,[23] and, independently, Siegbahn and Johannson,[24] have suggested possible angular momentum units for the decay scheme of $Mn^{56}$ which placed the ground state of $Fe^{56}$ at zero and the 0.822-Mev state of $Fe^{56*}$ at one. This value of $l=1$ falls within the $l$ values obtained from the half-life limit.

### E. $Sb^{122}$

The decay scheme of $Sb^{122}$ shows a single gamma-ray of 0.57 Mev as measured by Rall and Wilkinson,[25] by Kern, Zaffarano, and Mitchell,[26] and by Cook and Langer.[27]

The measurements on this state as plotted in Fig. 7 show a symmetric curve and consequently a half-life less than $1\times10^{-8}$ sec. Comparison with the Segrè-Helmholz formula places the $l$ value at two or less. Although internal conversion of the gamma-ray has been noted by Rall and Wilkinson,[25] the coefficient has not yet been measured. This measurement would provide an independent check on the $l$ value, but to date no correlation is possible.

### F. $Pr^{142}$

The exact decay scheme of $Pr^{142}$ is still subject to investigation, but there is reasonable agreement on a gamma-ray emission of low intensity and energy 1.5 to 2.2 Mev. Mandeville[28] has placed this gamma-ray energy at 1.74 Mev and coupled to a soft beta-spectrum (~0.215 Mev) with an occurrence about one-fiftieth of the strong 2.22 Mev beta-particle transition. This observation is reasonably consistent with other findings.

In this measurement both the proportional counter and a second polystyrene scintillation counter were used for beta-detection. In both cases the curve was symmetric, as shown in Fig. 8.

From the upper limit of $1\times10^{-8}$ sec. on the half-life, the Segrè-Helmholz formula gives an $l$ value of three or less, assuming a value of 1.74 Mev for the gamma-ray. No data for any other methods were available for correlation.

### G. $Au^{198}$

Measurement of the half-life of the 0.411 gamma-ray transition in the decay of $Au^{198}$ to $Hg^{198}$ has been previously reported by this writer.[29] The measured half-life of the excited state of $Hg^{198*}$ was placed at $2.3\times10^{-8}$ sec.

Since that time there have been a considerable number of conflicting reports on this state. Mandeville[30] reported that in his search for a possible metastable

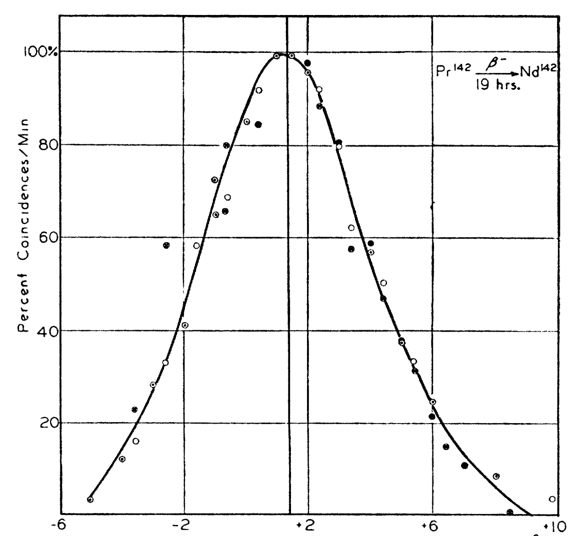


FIG. 8. Coincidence rate *versus* delay time curve for $Pr^{142}$ decay.

[24] K. Siegbahn and Johannson, Arkiv. Mat. Astron. Fys. **34A**, No. 10 (1947).
[25] W. Rall and R. G. Wilkinson, Phys. Rev. **71**, 321 (1947).
[26] Kern, Zaffarano, and Mitchell, Phys. Rev. **73**, 1142 (1948).
[27] C. S. Cook and L. M. Langer, Phys. Rev. **73**, 1149 (1948).
[28] C. E. Mandeville, Phys. Rev. **75**, 1257 (1949).
[29] W. J. MacIntyre, Phys. Rev. **76**, 312 (1949).
[30] C. E. Mandeville, private communication.

TABLE I.

| Isomer parent | Measured half-life (seconds) | Theoretical half-lives (Segrè-Helmholz formula) (seconds) | | | Indicated $l$ value from half-life | Indicated $l$ value from other measurements | Probable $l$ assignment |
|---|---|---|---|---|---|---|---|
| $Mg^{24*} \leftarrow Na^{24}$ | $>1\times10^{-8}$ | $l=2$ $6\times10^{-13}$ | $l=3$ $2\times10^{-8}$ | $l=4$ $1\times10^{-4}$ | $l\leq2$ | $l\geq2$ [a] | $l=2$ |
| $A^{38*} \leftarrow Cl^{38}$ | $>1\times10^{-8}$ | $l=2$ $2\times10^{-13}$ | $l=3$ $2\times10^{-9}$ | $l=4$ $1\times10^{-5}$ | $l\leq3$ | $l\leq2$ [b] | $l\leq2$ |
| $Ca^{42} \leftarrow K^{42}$ | $>2\times10^{-8}$ | $l=2$ $2\times10^{-13}$ | $l=3$ $3\times10^{-9}$ | $l=4$ $1\times10^{-5}$ | $l\leq3$ | $l=2$ [c] | $l=2$ |
| $Fe^{56*} \leftarrow Mn^{56}$ | $>1\times10^{-8}$ | $l=2$ $3\times10^{-12}$ | $l=3$ $1\times10^{-7}$ | | $l\leq2$ | $l=1$ [d] | $l=1$ |
| $Te^{122*} \leftarrow Sb^{122}$ | $>1\times10^{-8}$ | $l=2$ $6\times10^{-12}$ | $l=3$ $4\times10^{-7}$ | | $l\leq2$ | ... | $l\leq2$ |
| $Nd^{142*} \leftarrow Pr^{142}$ | $>1\times10^{-8}$ | $l=3$ $1\times10^{-10}$ | $l=4$ $1\times10^{-7}$ | | $l\leq3$ | ... | $l\leq3$ |
| $Hg^{198*} \leftarrow Au^{198}$ | $2.3\times10^{-8}$ | $l=2$ $1.7\times10^{-11}$ | $l=3$ $1.4\times10^{-6}$ | | $l=2\sim3$ | $l=2\sim3$ [e] $l=2$ [f] | $l=2$ |
| $Hg^{199*} \leftarrow Au^{199}$ | $>2\times10^{-8}$ | $l=2$ $1\times10^{-9}$ | $l=3$ $5\times10^{-7}$ | | $l\leq2$ | ... | $l\leq2$ |

[a] See references 17 and 18.
[b] See references 19 and 20.
[c] See reference 22.
[d] See references 23 and 24.
[e] D. Saxon and R. Heller, Phys. Rev. **75**, 909 (1949).
[f] D. Saxon, private communication.

state in this decay by looking for a loss in beta-gamma coincidences from $Au^{198}$ by reduction of resolving time of their coincidence circuit from 1.0 microsecond to $0.035\pm0.0002$ $\mu$sec., a decrease was noticed at the lower limit. This observation indicating a barely measurable lifetime for the $Hg^{198*}$ state, but their statistical limit was too great for an accurate estimate.

Bell and Petch[31] attempted measurement of this state with negative results. With their equipment a completely symmetrical curve was plotted, from which an upper limit of $3\times10^{-9}$ sec. was obtained. Deutsch and Wright[5] also obtained negative results and placed an upper limit of $4\times10^{-9}$ sec. on this state.

Recently, Jastram, Konneker, and Cleland[32] measured this state at $(4\pm1)\times10^{-8}$ sec., which is in fair agreement with this writer's results. Correlation with the Segrè-Helmholz formula has been previously reported.[28]

### H. $Au^{199}$

The decay of $Au^{199}$ involves only one beta-particle of energy, about 0.32 Mev, and several gamma-rays, as reported by Beach, Peacock, and Wilkinson,[33] and by Meem and Maienschein,[34] with the strongest gamma-ray about 0.230 Mev.

The coincidence curve taken on this decay gave poor resolution and poor statistical accuracy. The obtained curve was roughly symmetric, however, and no measurable half-life found. The upper limit of half-life was estimated at less than $2\times10^{-8}$ sec. This placed the transition of the strongest gamma-ray at $l$ equal to or less than two by correlation with the Segrè-Helmholz formula.

A summary of all the above results with prediction of $l$ values from the Segrè-Helmholz formula, correlation with $l$ values obtained from other methods, and probable $l$ assignments is shown in Table I.

The author wishes to express his thanks to Professor Ernest C. Pollard for valuable discussions throughout, and to Professor Howard L. Schultz for numerous discussions on equipment design and coincidence techniques.

[31] R. E. Bell and H. E. Petch, Phys. Rev. **76**, 1409 (1949).
[32] Jastram, Konneker, and Cleland, Phys. Rev. **79**, 243 (1950).
[33] Beach, Peacock, and Wilkinson, Phys. Rev. **76**, 1585 (1949).
[34] J. L. Meem, Jr. and F. C. Maienschein, Phys. Rev. **76**, 328 (1949).