

## Lifetimes of Energy Levels in $\text{Al}^{28}$ , $\text{Mn}^{56}$ , $\text{Cu}^{64}$ , $\text{Rh}^{104}$ , and $\text{I}^{128}$ Excited by Slow Neutron Capture\*

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Measurements of delayed coincidences between  $\gamma$  rays were used to determine the half-lives of states excited by slow-neutron capture in various nuclei. The half-lives found were:  $\text{Mn}^{56}$ , first excited level  $11.4_{-3}^{+2}$  nsec, second excited level  $5.1 \pm 0.5$  nsec;  $\text{Rh}^{104}$ , first excited level  $2.6 \pm 0.2$  nsec, 96-keV transition  $\leq 0.6$  nsec;  $\text{Cu}^{64}$ , first excited level  $\leq 0.3$  nsec, second excited level  $\leq 0.3$  nsec;  $\text{Al}^{28}$ , first excited level  $2.3 \pm 0.2$  nsec;  $\text{I}^{128}$ , 30-keV level  $8.8 \pm 1.0$  nsec, 137-keV level  $8.0 \pm 0.6$  nsec, 90-keV transition  $\leq 0.7$  nsec. Measurements of pulse-height spectra satisfying the condition of prompt and delayed coincidence and measurements at neutron resonances of iodine were used to establish the presence of excited states at 30 and 137 keV in  $\text{I}^{128}$ . A previously unreported neutron resonance was observed in  $\text{I}^{127}$  at a neutron energy of 10.7 eV.

### I. INTRODUCTION

ABSORPTION of a slow neutron by a nucleus usually results in the prompt emission of a cascade of  $\gamma$  rays which appreciably populates low-lying states of both parities and various spins. The observation of the low-energy part of a capture- $\gamma$ -ray spectrum thus sometimes reveals transitions inaccessible by  $\beta$  decay because of spin considerations. In addition, many of the isotopes which can be excited by neutron capture, especially odd-odd nuclei, cannot be reached by  $\beta$  decay.

In spite of the fact that  $\gamma$  rays from capture of thermal neutrons in about 60 elements have already been studied, very few of these spectra, even among the light elements, are understood in detail.<sup>1</sup> This is especially true of the lifetimes of low-lying states, which depend on specific nuclear mechanisms—in contrast to internal conversion processes, which should not depend on the details of nuclear structure. Only one experiment on the lifetimes of excited states fed by neutron capture has, however, been reported in the literature. This was a determination of the half-lives of the 26-, 109-, and 210-keV levels in  $\text{Mn}^{56}$  by D'Angelo.<sup>2</sup>

The present paper describes work done on the determination of the half-lives for the 26- and 109-keV states in  $\text{Mn}^{56}$ , the 51-keV state and the state associated with a 96-keV transition in  $\text{Rh}^{104}$ , the 159- and 275-keV states of  $\text{Cu}^{64}$ , the 32-keV state of  $\text{Al}^{28}$ , and two states in  $\text{I}^{128}$ . The  $\gamma$ -ray spectra from neutron capture in these isotopes were also studied, and in some cases it was possible to obtain new data in connection with the energies of excited states.

### II. APPARATUS

In determining the half-lives of the excited states studied, samples of the elements investigated were

bombarded by a beam of thermal neutrons from the Argonne CP-5 reactor. Gamma rays coming from the sample were observed in delayed coincidences by two scintillators and photomultipliers. The pulses from the photomultipliers were simultaneously fed to fast and slow circuits. The fast circuitry measured the time difference between pulses coming from the two photomultipliers by means of a time-to-amplitude converter. The slow circuitry selected the specific events of interest, an integral bias being used in the one channel to select the high-energy  $\gamma$  rays from the capture events, whereas a differential pulse-height discriminator in the other channel selected pulses caused by  $\gamma$  rays of a specified low energy. The equipment used was essentially the same as that described by D'Angelo,<sup>2</sup> the circuit used for time-to-amplitude conversion being based on that developed by Green and Bell.<sup>3</sup>

To form a beam that consisted predominantly of subthermal neutrons, free of  $\gamma$  rays and fast neutrons, the beam hole in the CP-5 reactor was solidly plugged by lead cylinders having a total length of 49 cm. The beam was further filtered by passing it through 10 cm of bismuth and then defined by a collimator consisting of two layers of boral 6 mm thick alternated with two layers of lead, 5 cm thick. The samples used for irradiation were made to be approximately a half thickness for the specific  $\gamma$  ray to be observed, and were suspended from a light aluminum strip at an angle of about  $45^\circ$  to the beam direction.

A plastic scintillator 38 mm high with a diameter of 50 mm was viewed by an RCA-6342 photomultiplier to form a zero-time signal by detecting the high-energy capture  $\gamma$  rays; and a NaI(Tl) crystal 6 mm thick by 38 mm in diam, mounted on an RCA-6810A photomultiplier, detected the low-energy  $\gamma$  rays originating from the transition whose lifetime was to be determined. Although the technique used here is similar to the delayed-coincidence techniques used for studying the lifetimes of states excited by  $\beta$  decay, and although a plastic scintillator would give better time resolution than the NaI(Tl) crystal, the complexity of the  $\gamma$  spectra from

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<sup>1</sup> G. A. Bartholomew, in *Nuclear Spectroscopy*, edited by Fay Ajzenberg-Selove (Academic Press, Inc., New York and London, 1960), Part A, p. 306.

<sup>2</sup> N. D'Angelo, Phys. Rev. **117**, 510 (1960).

<sup>3</sup> R. E. Green and R. E. Bell, Nuclear Instr. **3**, 127 (1958).

slow-neutron capture demands the higher energy resolution of a NaI(Tl) scintillator.

To obtain favorable coincidence counting rates, the scintillation counters were placed as close together as possible without intercepting the beam. The scintillation counters were screened from general background by a minimum of 10 cm of lead enclosed in boral and cadmium. The neutron beam passed through an opening in the back of the shielding and into a beam catcher. Care was taken to suspend the samples with a minimum width of masking tape to reduce neutron scattering as much as possible. The disappearance of the prominent 137-keV line of  $I^{128}$  from the spectrum observed by the NaI(Tl) crystal was used as a criterion for determining whether neutron scattering had been reduced to acceptable limits. An internal gate on the reactor was used to adjust the intensity of the neutron beam so that the counting rates in the separate channels did not overload the amplifiers and discriminators.

It was found that the 6BN6 tubes used in the time-to-amplitude converter<sup>3</sup> showed fairly wide variations in characteristics from tube to tube. The characteristics of twenty tubes were determined and the two tubes showing the best characteristics were selected for this work. The pulses from the converter were amplified and recorded on a transistorized 200-channel analyzer.

Any maladjustment of the timing of pulses arriving at the diode supervisory coincidence circuit<sup>3</sup> with respect to that of pulses arriving at the 6BN6 may cause distortion of the delay curve at either end. The time calibration of the time-to-amplitude converter also changes with aging of the 6BN6 tube. To eliminate systematic errors that might be introduced by these factors, a time calibration was done after each run by shortening the cable, marked 287 cm in the circuit of Green and Bell,<sup>3</sup> in at least 5 successive steps to cover the whole time range of the circuit. This could be done in our case because this cable had to be made 1236 cm long to compensate for the greater delay in the 6810A photomultiplier compared to that of the 6342 photomultiplier used in the "prompt" channel. Changing the length of this cable has the effect of introducing the same artificial delay for pulses arriving from the 6810A photomultiplier at both the diode coincidence circuit and the 6BN6 circuit and, in addition to supplying a time calibration, also serves as a check on the time linearity of the circuit.

### III. RESULTS

#### A. Manganese-56

The functioning of the equipment was checked by repeating D'Angelo's measurements of the half-lives of the first two excited levels of  $Mn^{56}$ . First the spectrum of  $Mn^{56}$  was observed with the NaI(Tl) crystal and found to be identical to that observed by D'Angelo,<sup>2</sup> including the line at 137 keV which he correctly ascribed to neutron capture in the iodine of the NaI(Tl) crystal.

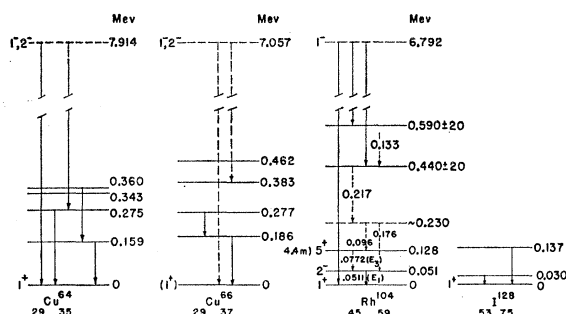


FIG. 1. Level schemes of  $Cu^{64}$ ,  $Cu^{66}$ ,  $Rh^{104}$ , and  $I^{128}$ .

It was, however, possible to eliminate this line completely by eliminating from the sample mounting all hydrogenous material that could scatter neutrons.

Half-lives of  $11.4_{-3}^{+2}$  nsec and  $5.1 \pm 0.5$  nsec were found for the first and second excited states of  $Mn^{56}$ , respectively, in agreement with the results of D'Angelo.<sup>2</sup> The result for the first excited level is significant because it is free of a systematic uncertainty which may be present in D'Angelo's result for this state. A contribution from the  $I^{128}$  spectrum is contained in D'Angelo's measurement, as is shown by the presence of the 137-keV line in the spectrum observed by him. In the present work it was found that  $I^{128}$  has a line at 30 keV with a half-life of 8.8 nsec, which may have influenced the previous determination of the half-life of the 26-keV transition in  $Mn^{56}$ . However, since the 137-keV line was completely eliminated from our spectrum, the half-life determination of the Mn transition was not influenced by contributions from  $I^{128}$  in the present measurement.

#### B. Rhodium-104

The  $\gamma$ -ray spectrum of  $Rh^{104}$ , when excited by neutron capture in  $Rh^{103}$ , has been studied by a number of workers.<sup>4-9</sup> The results of this work are summarized in the decay scheme for the low-lying states of  $Rh^{104}$  shown in Fig. 1. The scheme shown is based on that given in the Nuclear Data Sheets<sup>10</sup> and by Estulin *et al.*<sup>8</sup>

A 0.112-g/cm<sup>2</sup> rhodium sample was made of powdered rhodium mixed with sulfur and placed in an aluminum can with a wall thickness of 4.5 mg/cm<sup>2</sup>. The spectrum

<sup>4</sup> B. Hamermesh and V. Hummel, Phys. Rev. **88**, 916 (1952).

<sup>5</sup> G. A. Bartholomew and B. B. Kinsey, Can. J. Phys. **31**, 1025 (1953).

<sup>6</sup> L. V. Groshev, V. N. Lutsenko, A. M. Demidov, and V. I. Pelekhov, *Atlas of  $\gamma$ -Ray Spectra from Radiative Capture of Thermal Neutrons* (Pergamon Press, New York, 1959).

<sup>7</sup> I. V. Estulin, L. F. Kalinkin, and A. S. Melioranskii, J. Exptl. Theoret. Phys. (U.S.S.R.) **31**, 886 (1956) [translation: Soviet Phys.—JETP **4**, 752 (1957)].

<sup>8</sup> I. V. Estulin, L. F. Kalinkin, and A. S. Melioranskii, J. Exptl. Theoret. Phys. (U.S.S.R.) **32**, 979 (1957) [translation: Soviet Phys.—JETP **5**, 801 (1957)].

<sup>9</sup> J. E. Draper, Phys. Rev. **114**, 268 (1959).

<sup>10</sup> *Nuclear Data Sheets*, National Academy of Sciences—National Research Council, 1960 (U. S. Government Printing Office, Washington, D. C.).

observed with the NaI(Tl) crystal during neutron bombardment showed the same lines observed by Draper,<sup>9</sup> i.e., at about 51, 96, 131, 185, and 220 keV. The 51-keV transition, which presumably results from the decay of the first excited state of  $\text{Rh}^{104}$ , showed up even more prominently than in Draper's spectrum.

The 51- and 96-keV lines were chosen for study. Decay curves were taken by consistently requiring that the "prompt"  $\gamma$  ray spend more than 400 keV of its energy in the plastic scintillator and that the energy lost in the NaI(Tl) by the "delayed"  $\gamma$  ray be within a window that lets through the peak at either 96 or 51 keV, the limits of the window being set to pass the peak out to half its maximum height on each side.

The time-distribution curves obtained were compared to prompt resolution curves obtained by replacing the rhodium sample by a sample of mercury (in the form of  $\text{Hg}_2\text{S}$ ), a material for which the high-energy  $\gamma$ -ray spectrum is similar to that of rhodium and for which almost all of the  $\gamma$  rays are expected to be emitted with a negligible delay. Mercury was chosen because the isotope  $\text{Hg}^{199}$  is responsible for almost 100% of the neutron capture and the  $\text{Hg}^{200}$  formed is a nuclide about which we have considerable information.<sup>6,11</sup> In particular, the low-energy spectrum is dominated by a 368-keV  $\gamma$  ray with an intensity of 40 quanta per 100 neutrons captured. The half-life of the first excited state responsible for this transition is known to be about  $6 \times 10^{-11}$  sec, which is effectively "prompt" for our purpose. Since the 368-keV transition is from a  $2^+$  state to the  $0^+$  ground state, and since most of the transitions from higher states would be of a similar character but of higher energy, one has every reason to believe that the time delay for these transitions would be even shorter. Other elements for which most of the capture  $\gamma$  radiation is expected to be prompt are cadmium and platinum. These materials were used instead of mercury when it was desirable to have a different high-energy  $\gamma$ -ray spectrum.

Although the mercury sample gave no resolved lines near the energies of the two rhodium lines investigated, there were sufficient counts from the Compton tail of high-energy  $\gamma$  rays to ensure a reasonable coincidence counting rate when the single-channel rates were kept the same as in the case of rhodium. This was done consistently because the functioning of the electronic circuits used for selecting the energy on the delayed side was to a certain extent dependent on the singles counting rate in this channel.

The decay curve obtained for the 51-keV transition to the ground state of  $\text{Rh}^{104}$  is shown as curve A in Fig. 2. For comparison, the "prompt" resolution curve obtained with mercury is shown as curve B. The number of counts/channel refers to curve A and is in arbitrary units for curve B.

As the time-resolution curve at this energy is fairly

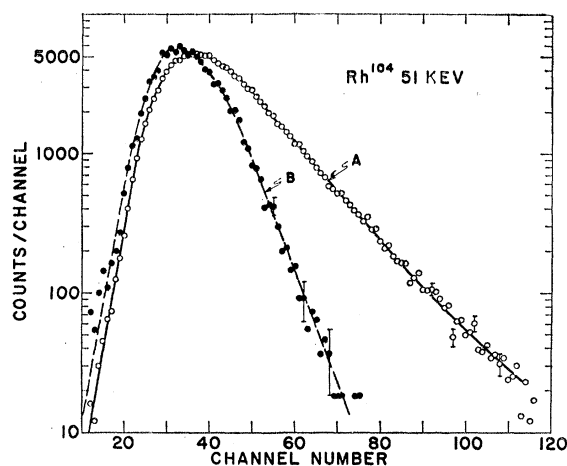


FIG. 2. Decay curve for 51-keV transitions in  $\text{Rh}^{104}$  (curve A). Curve B shows the prompt time-resolution curve taken with mercury. Counts/channel for curve B in arbitrary units.

broad, a calculation was made to ascertain the influence of resolution broadening on the shape of the measured decay curve. The calculation consisted of the evaluation of the equation<sup>12</sup> describing the convolution of the prompt resolution curve with the decay function  $f(t)$ , namely,

$$d_f(t) = \int_0^\infty f(t')P(t-t')dt', \quad (1)$$

where  $d_f(t)$  is the curve to be expected for a state decaying according to  $f(t)$  and  $P(t)$  is the resolution curve. The prompt curve was assumed to be a Gaussian distribution having a width determined from only the right-hand side of the experimental resolution curve, since instrumental cutoff may cause a deformation of the left-hand side. The function  $f(t)$  was taken as  $e^{-t/\tau}$  and the value of  $\tau$  was determined from the straight portion of curve A. Under these assumptions the integration showed that  $d_f(t)$  becomes a pure exponential decay with a mean life  $\tau$  beyond about channel 50. Consequently, only data points for channel numbers greater than 50 were used for evaluating the experimental half-life.

It will be noted that curve A (Fig. 2) deviates slightly from the pure exponential shape above channel 85. This occurred because a slight malfunctioning of the diode supervisory circuit<sup>3</sup> allowed a small number of pulses of more than maximum overlap in the 6BN6 circuit to slip through.

Using only channels 50 to 85, one obtains a half-life of  $2.6 \pm 0.2$  nsec for the first excited level of  $\text{Rh}^{104}$ . The uncertainty given is an estimate of the possible magnitude of systematic errors and is not determined by statistics.

In the case of the 96-keV transition, the decay curve and the prompt curve were of similar shape over a total

<sup>11</sup> D. Strominger, J. M. Hollander, and G. T. Seaborg, *Revs. Modern Phys.* **30**, 585 (1958).

<sup>12</sup> Z. Bay, *Phys. Rev.* **77**, 419 (1950).

of 3.5 decades, except for a small shift probably due to instability in the apparatus. This places an upper limit of 0.6 nsec on the half-life of this transition, the limit being set by the stability of the equipment.

According to an argument given in the Nuclear Data Sheets,<sup>10</sup> the 51-keV excited state of Rh<sup>104</sup> is characterized by spin 2 and negative parity. Hence, the state is expected to decay to the 1<sup>+</sup> ground state by an *E1* radiative transition. Because of the scarcity of experimentally determined widths of low-energy *E1* transitions for heavy nuclides, it is of interest to compare the width for the 51-keV transition of Rh<sup>104</sup> with the Weisskopf estimate.<sup>13</sup> The mean lifetime  $\tau_\gamma$  for the radiative transition is  $\tau_\gamma = (1+\alpha)\tau$ , where  $\alpha$  is the total internal conversion coefficient. Using a value  $\alpha=0.95$ , as given by Rose,<sup>14</sup> we obtain  $\tau_\gamma=7.4$  nsec. This value corresponds to a width of  $9.0 \times 10^{-8}$  ev. Hence, making use of the Weisskopf estimate in the convenient form given by Wilkinson,<sup>15</sup> one finds that the ratio of the experimental width to the Weisskopf estimate is  $3 \times 10^{-4}$ , a value which indicates that the transition is greatly retarded. However, when the number  $3 \times 10^{-4}$  is compared with the values for other *E1* transitions in heavy nuclides, as summarized by Wilkinson,<sup>15</sup> one observes that the *E1* transition for Rh<sup>104</sup> is not abnormally slow. Indeed, it is one of the fastest *E1* transitions yet reported for the decay of a low-energy state of a heavy nuclide.

### C. Copper-64

Using the  $\text{Cu}^{63}(d,p)\text{Cu}^{64}$  and  $\text{Cu}^{65}(d,p)\text{Cu}^{66}$  reactions on separated isotopes, de Figueiredo *et al.*<sup>16</sup> found excited states at  $159 \pm 8$  and  $277 \pm 8$  keV in  $\text{Cu}^{64}$  and at  $183 \pm 8$  and  $272 \pm 8$  keV in  $\text{Cu}^{66}$ . The low-energy gamma spectra of  $\text{Cu}^{64}$  and  $\text{Cu}^{66}$  bombarded by thermal neutrons were studied by Skliarevskii *et al.*,<sup>17</sup> who also used separated isotopes of copper. In the  $\text{Cu}^{64}$  spectrum they observed lines at  $155 \pm 5$ ,  $205 \pm 10$ , and  $276 \pm 10$  keV; and in the  $\text{Cu}^{66}$  spectrum lines were observed at  $92 \pm 5$  and  $180 \pm 10$  keV. Except for the 205-keV transition, these lines can be explained as due to transitions between the low-lying states found by de Figueiredo *et al.*,<sup>16</sup> as shown in Fig. 1. All five lines, but with somewhat different energies, were also found by Urbanec *et al.*<sup>18</sup> in the neutron-capture  $\gamma$ -ray spectrum of natural

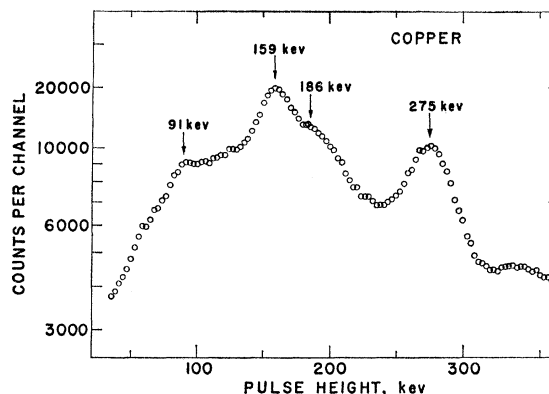


FIG. 3. Singles spectrum of capture  $\gamma$  rays from natural copper.

copper. The lines at 137, 88, and 33 keV in their published spectrum are presumably due to neutron capture in the NaI(Tl) crystal since, as seen in Fig. 3, these lines do not appear in the spectra measured in the present experiment. Our spectrum was taken with a  $10 \times 10$  cm NaI(Tl) crystal well shielded from neutrons by using a  $\text{B}_4\text{C}$  filter between the copper sample and the detecting crystal.

As Fig. 3 shows, the only lines that appear prominently in the spectrum of normal copper are those at  $91 \pm 4$  and  $186 \pm 5$  keV from  $\text{Cu}^{66}$  and at  $159 \pm 2$  and  $275 \pm 3$  keV from  $\text{Cu}^{64}$ . The arrangement of the low-lying levels in  $\text{Cu}^{64}$  and  $\text{Cu}^{66}$  (Fig. 1) was taken from reference 6, except that the energies of the two lowest states in each case were altered to be consistent with the values of the  $\gamma$ -ray energies found in this work. In the case of  $\text{Cu}^{64}$ , at least, the energies derived in this way are probably more accurate than those given by the  $(d,p)$  measurements.<sup>16</sup>

Delay runs were done on the 275- and 159-keV transitions in  $\text{Cu}^{64}$ . The time curve obtained for the 275-keV line was compared to a prompt curve obtained by bombarding a platinum foil in place of the copper sample. The prompt curves and delay curves looked alike and allowed an upper limit of 0.3 nsec to be placed on the half-life of the second excited level of  $\text{Cu}^{64}$ .

The decay curve taken for the 159-keV transition in  $\text{Cu}^{64}$  was compared with prompt curves obtained by irradiating platinum, mercury, and cadmium. A total of 4 delay curves and 8 prompt curves showed the same form, with maximum centroid shifts equivalent to a half-life of 0.3 nsec. As the shift was not reproducible, this must be ascribed to instrumental instability and allows an upper limit of 0.3 nsec to be set on the half-life of the lowest level of  $\text{Cu}^{64}$ .

### D. Aluminum-28

The first excited state of Al<sup>28</sup> is at 32 keV,<sup>11</sup> and a 32-keV line was the only low-energy line observed in the capture  $\gamma$  spectrum of an aluminum source exposed to thermal neutrons. The NaI(Tl) crystal was well

<sup>13</sup> V. F. Weisskopf, Phys. Rev. **83**, 1073 (1951).

<sup>14</sup> M. E. Rose, in *Nuclear Spectroscopy*, edited by Fay Ajzenberg-Selove (Academic Press Inc., New York and London, 1960), Part B, p. 834.

<sup>15</sup> D. H. Wilkinson, in *Nuclear Spectroscopy*, edited by Fay Ajzenberg-Selove (Academic Press Inc., New York and London, 1960), Part B, p. 852.

<sup>16</sup> R. P. de Figueiredo, M. Mazari, and W. W. Buechner, Phys. Rev. **112**, 873 (1958).

<sup>17</sup> V. V. Skliarevskii, E. P. Stepanov, and B. A. Obiniakov, Atomnaya Energiya **5**, 454 (1958), [translation: Soviet Journal of Atomic Energy **5**, 1350 (1958)].

<sup>18</sup> J. Urbanec, J. Kajfosz, and J. Kopecký, Czechoslov. J. Phys. **10**, 275 (1960).

shielded from thermal neutrons by means of a  $B_4C$  filter.

Figure 4 shows the decay curve of the first excited state, curve A, and a prompt curve B obtained by using the mercury sample. The data were analyzed as described for the 51-keV transition in  $Rh^{104}$ . This analysis showed that the shape of the decay curve is not influenced by the width of the prompt resolution curve above channel 30. A weighted least-squares fit to the data above channel 30 gave a half-life of  $2.32 \pm 0.03$  nsec for the decay of this state. Observed changes in the position of the prompt resolution peak during the course of these runs lead us to assign a wider margin of error and to give the half-life as  $2.3 \pm 0.2$  nsec. This value agrees within the experimental error with the value  $2.1 \pm 0.3$  nsec found by Severiens and Hanna,<sup>19</sup> who used a recoil technique.

### E. Iodine-128

Although a number of papers have reported information about the low-energy  $\gamma$  rays from neutron capture in  $I^{127}$ , the data are incomplete and in some cases contradictory. Reier and Shamos<sup>20</sup> found a weak line at 255 keV and indications of a very intense capture  $\gamma$  ray at about 85 keV. Balzer *et al.*<sup>21</sup> found low-energy lines at  $260 \pm 15$  and  $130 \pm 10$  keV, the intensity of the 130-keV line being no bigger than that of the 260-keV line. Draper,<sup>9</sup> on the other hand, found a very prominent 135-keV line and evidence of only weak transitions at 80 keV and in the 240–300 keV region, in substantial agreement with Estulin *et al.*<sup>7,8</sup> who, however, did not

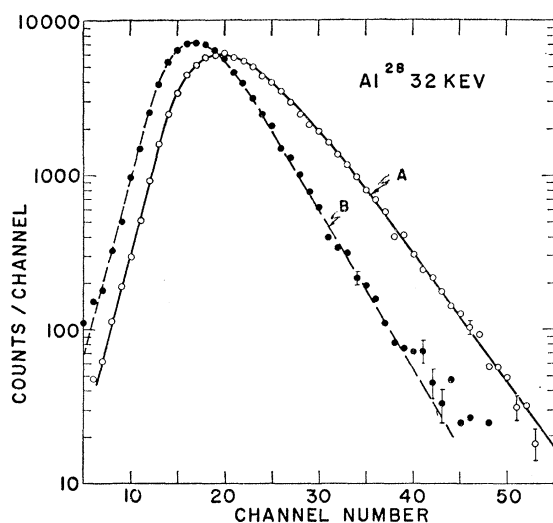


FIG. 4. A. Decay curve for 32-keV transitions in  $Al^{28}$ . B. Prompt time-resolution curve using mercury as source. Counts/channel in arbitrary units for curve B.

<sup>19</sup> J. C. Severiens and S. S. Hanna, Phys. Rev. **104**, 1612 (1956).  
<sup>20</sup> M. Reier and M. H. Shamos, Phys. Rev. **100**, 1303 (1955).  
<sup>21</sup> R. Balzer, H. Knoepfel, J. Lang, R. Müller, and P. Stoll, Nuovo cimento **11**, 609 (1959).

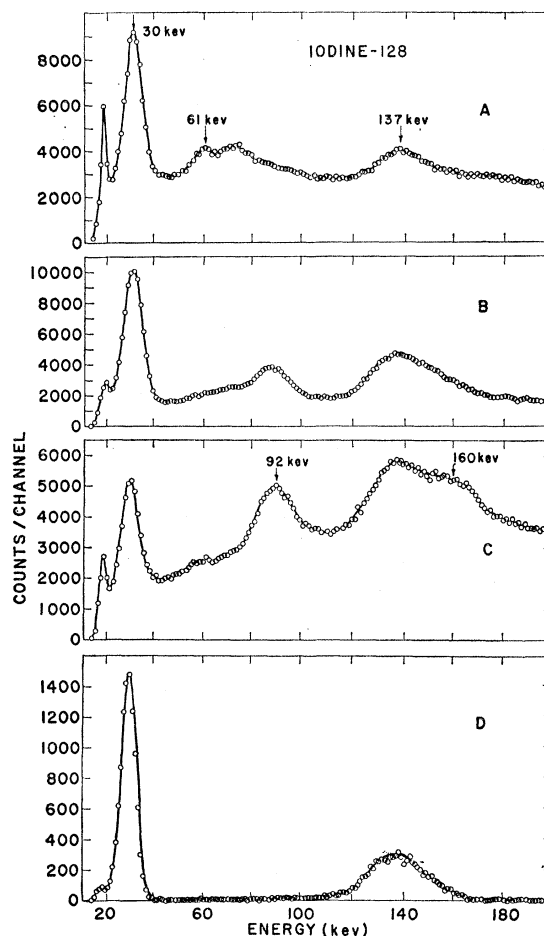


FIG. 5. Neutron-capture  $\gamma$ -ray spectra of  $I^{128}$  taken with NaI(Tl) crystal 6 mm thick. A. Singles spectrum with external sample. B. Singles spectrum with detector crystal as target. C. Prompt coincidence spectrum with crystal as target. D. Delayed coincidence spectrum with detector crystal as target.

observe a line at 80 keV when their counter was shielded with  $B_4C$ . In subsequent work, Kalinkin *et al.*<sup>22</sup> resolved the 135-keV peak and found  $I^{128}$  lines at  $28 \pm 2$ ,  $135 \pm 3$ , and  $158 \pm 4$  keV with 23, 20, and 7.5 gamma quanta, respectively, emitted per 100 neutrons captured in  $I^{127}$ . The line at 28 keV was ascribed to characteristic  $K$  radiation caused by internal conversion of the  $\gamma$  rays from the  $I(n, \gamma)$  reaction.

In the course of this investigation it was found that the decay of  $I^{128}$  following neutron capture is rather more involved than had so far been recognized. The low-energy capture- $\gamma$ -ray spectrum of a 100 mg/cm<sup>2</sup> sample of NaI irradiated by thermal neutrons is shown in Fig. 5, curve A. NaI was used for the target because it is in a convenient chemical form. Sodium is expected to contribute little to the observed spectrum because the thermal-neutron-capture cross section for iodine is

<sup>22</sup> L. F. Kalinkin, A. S. Melioranskii, and I. V. Estulin, J. Exptl. Theoret. Phys. (U.S.S.R.) **36**, 1613 (1959) [translation: Soviet Phys.—JETP **9**, 1146 (1959)].

almost 13 times as large as for sodium. A NaI(Tl) crystal 6 mm thick was used as detector and was well screened from scattered neutrons by using a B<sub>4</sub>C filter. Prominent lines appeared at 137 and 30 keV. The width and asymmetry of the 137-keV line indicates that it consists of at least two lines, in agreement with the results found by Kalinkin *et al.*<sup>22</sup> There is, however, no evidence of the line at 80 keV observed by Draper.<sup>9</sup> The lines at 61 and about 70 keV are probably caused by background radiations.

Figure 5, curve B, shows the same spectrum, but this time the 6-mm crystal itself was used as target in the thermal neutron beam. Again the lines at 137 and 30 keV appear, with the relative intensities altered in favor of the 137-keV line by a factor of almost two. A line observed at about 90 keV under these conditions is discussed later.

Since the 137-, 90-, and 30-keV lines are the most prominent in the  $\gamma$  spectrum, the window of the differential discriminator in the "delayed" channel of the time-measuring equipment was set on these lines, and the half-lives were determined. Figure 6 shows the time distribution for the 137-keV decay. The sample used was the 100-mg/cm<sup>2</sup> NaI sample, and the NaI(Tl) scintillator itself was well shielded from scattered neutrons by a B<sub>4</sub>C filter. The decay curve in Fig. 6 is actually a composite of separate runs with various delays introduced in the prompt channel to enable the curve to be followed for longer delay times. The prompt resolution curve, given for comparison, was obtained by using the capture  $\gamma$  rays from cadmium as a source of prompt coincidences.

Figure 6 shows that the decay is complex, consisting of a part that is approximately prompt and a part with a half-life of  $8.0 \pm 0.6$  nsec. By extrapolating the exponential portion in both directions, deducting the

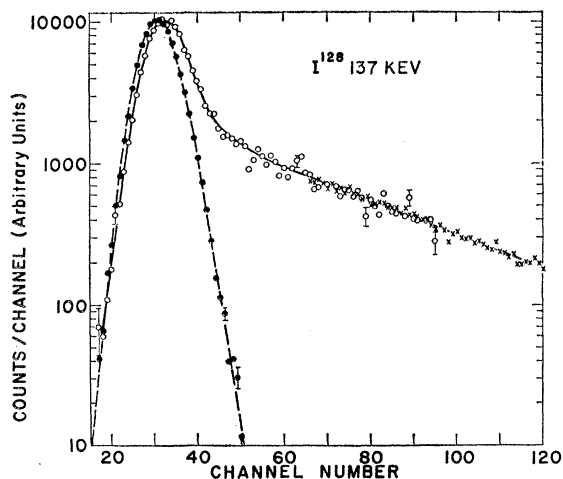


FIG. 6. Decay of 137-keV transitions in  $I^{128}$  (open circles and crosses). Artificial delay was introduced to obtain tail of the decay curve. The prompt time-resolution curve (filled circles) was obtained by using capture  $\gamma$  rays from cadmium.

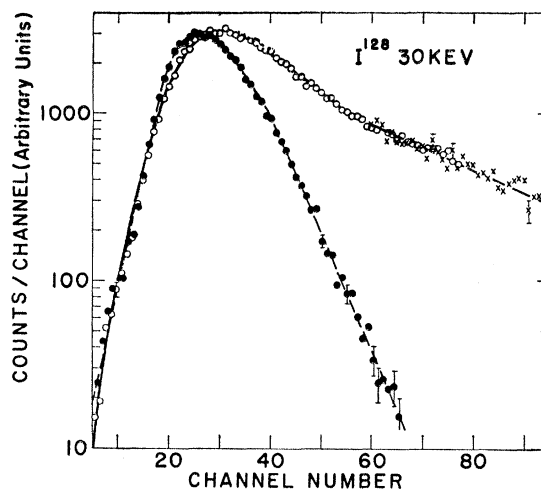


FIG. 7. Decay of 30-keV transitions in  $I^{128}$  (open circles and crosses). Artificial delay was introduced to obtain the tail of the decay curve. The prompt time-resolution curve (filled circles) was obtained by using capture  $\gamma$  rays from mercury.

extrapolated curve from the experimental curve, and using the form of the prompt time-resolution curve as a guide, it is possible to estimate the ratio of prompt to delayed decays as 1.4:1.

To obtain a favorable coincidence counting rate for the 30-keV transition, the 6 mm NaI(Tl) crystal was used as target. The decay curve is shown in Fig. 7 together with a resolution curve obtained by irradiating mercury. In this case, too, the decay is complex, showing a prompt component and a component with a half-life of  $8.8 \pm 1.0$  nsec. The ratio of prompt to delayed decays is, however, much smaller, being 0.4:1 in this case.

The line at 90 keV was investigated, again with the NaI(Tl) crystal as target. The time-delay curve obtained was effectively "prompt" and allowed an upper limit of 0.7 nsec to be placed on the half-life of this transition.

It could be argued that the prompt component in the 137- and 30-keV decays was due to the prompt Compton base on which the lines are superimposed. To test this the output of the time-to-amplitude converter was fed to a differential discriminator, the output of which was used to trigger the 200-channel analyzer. Pulses from the NaI(Tl) crystal were then fed to the analyzer, the crystal again being used as target. First the window of the differential discriminator was set on the prompt peak of the 6BN6 output. The resultant spectrum is shown in Fig. 5, curve C. The discriminator was then set to accept all 6BN6 pulses above the prompt peak and Fig. 5, curve D, was recorded. This curve shows that only the 137- and 30-keV lines contribute delayed coincidences with delays above  $\approx 12$  nsec. Similar results were found when using the external NaI target and screening the crystal from neutrons. At the same time the 137-keV line appears prominently in the prompt coincidence spectrum C, whereas the relative

intensity of the 30-kev line is greatly reduced. Even if we assume that the 30-kev decay is a pure exponential with a half-life of 8.8 nsec and that the presence of the 30-kev peak in spectrum C results from the earliest part of this decay, we are forced to conclude that the 137-kev transition shows a composite decay, one component having a very short half-life and a comparable fraction having a half-life of 8 nsec. It may be pointed out that this technique of obtaining a delayed coincidence spectrum can be used to obtain a quick indication of which lines in a spectrum contribute  $\gamma$  rays with a measurable delay, before studying the decay of the specific  $\gamma$  transition in detail.

As expected, the 90-kev line appears in the prompt coincidence spectrum C. In addition, curve C emphasizes the presence of a line at about 160 kev, although lack of resolution and uncertainty about the number of  $\gamma$ -ray lines that form the observed peak still make it difficult to obtain an accurate determination of the energy. The line at 160 kev may be identified with the  $158 \pm 4$  kev line reported by Kalinkin *et al.*<sup>22</sup>

In an effort to understand the origin of the lines for which delayed time distributions were measured, the pulse-height spectra from capture of neutrons in NaI were studied under a wide variety of conditions. One of the first problems that needed to be solved was the identification of the nuclide responsible for each line. To help obtain this information, the Argonne fast chopper<sup>23</sup> was used to study the pulse-height spectra that result from capture of neutrons in the resonances of iodine. At these resonances the neutron-capture cross section of iodine is so high that, even when the target is a NaI(Tl) scintillator, one can be sure that almost all of the neutron capture takes place in iodine.

The target used in the measurements of the spectra

for resonant capture was a 5×5 cm NaI(Tl) scintillator. The electronic system employed to select the neutron time-of-flight corresponding to the desired neutron energy and to record the pulse-height spectra has been described by Carpenter and Bollinger.<sup>24</sup> This system was used in measurements with neutrons of 8 different energies within the range from 8 to 30 ev and also for neutrons of thermal energy.

The singles spectrum obtained at the well-known resonance at 20 ev is given in Fig. 8. In these data a correction was made for the contribution from background radiations, as measured at a slightly higher energy than the resonance. The most significant features of the spectrum are the lines at about 30 and 90 kev and the system of lines beginning at about 137 kev. These same lines, with the same relative intensities, were also observed at a previously unreported resonance at a neutron energy of about 10.7 ev. For thermal neutrons, the spectrum is likewise similar to Fig. 8, insofar as the lines at 30 and 137 kev are concerned, but the 90-kev line is about twice as strong as the corresponding line observed at the neutron resonances.

In Fig. 8 we observe that, in addition to the prominent lines at  $30 \pm 1$ ,  $88 \pm 2$ , and  $137 \pm 2$  kev, there is evidence for lines at  $182 \pm 5$ ,  $220 \pm 5$ ,  $240 \pm 5$ ,  $300 \pm 5$ ,  $390 \pm 10$ , and  $430 \pm 10$  kev. Also, the asymmetry of the peak at 137 kev again indicates that it is formed by at least two  $\gamma$  rays. The spectrum from capture of thermal neutrons also has structure which indicates the presence of lines at approximately the energies given above.

The close similarity of the spectra from capture of neutrons at thermal and resonance energies indicates that, for the thermal neutrons also, a large part of the intensity of the lines at 30 and 137 kev results from capture in iodine. The greater relative intensity of the 90-kev line for thermal neutrons might be explained in one of two alternative ways. Either some source other than  $I^{128}$  also contributes to the 90-kev line in the thermal spectra or the intensity of the 90-kev line, which might be formed by the summing of pulses, depends on the proximity of the position of neutron capture to the surface of the scintillator. The latter interpretation must be rejected because the relative intensity of the 90-kev line is found to be the same in off-resonance regions (such as at a neutron energy of 15 ev), where the capture takes place in the interior of the crystal, as it is at the resonance, where neutrons are captured at the surface. Thus we conclude that the peak at about 90 kev in the thermal spectrum is formed by lines from two independent sources. This conclusion is reinforced by the observation that the energy of  $92 \pm 1$  kev for the line in the thermal spectrum differs significantly from the energy of  $88 \pm 2$  kev for the line in the spectrum for resonant capture.

One would expect the additional source for the 90-kev

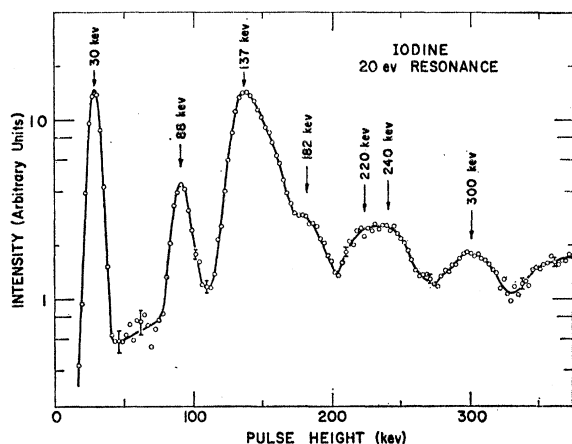


FIG. 8. Singles spectrum of  $I^{128}$ , obtained with a NaI(Tl) crystal used as target at the 20-ev resonance

<sup>23</sup> L. M. Bollinger, R. E. Coté, and G. E. Thomas, *Proceedings of the Second International Conference on Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1958), Vol. 14, p. 239.

<sup>24</sup> R. T. Carpenter and L. M. Bollinger, *Nuclear Phys.* **21**, 66 (1960).

line to be capture in Na. This conjecture tends to be confirmed by the results of a measurement of the spectrum from neutron capture in a target of  $\text{NaNO}_3$ , in which Na is the only nuclide that is expected to give low-energy gamma rays. The only line observed was a weak one at  $93 \pm 3$  kev. This line probably results from a decay from the second to the first excited state of  $\text{Na}^{24}$ , for which states the reported difference<sup>10</sup> in energy is  $92 \pm 12$  kev. If so, our measurement of the decay curve for the 92-kev line from a NaI target sets a limit of 0.7 nsec to the lifetime of the 564-kev state of  $\text{Na}^{24}$  as well as to some as yet unknown state of  $\text{I}^{128}$ . According to the above argument, some influence of the Na  $\gamma$  ray must be present in spectrum A of Fig. 5. However, the background contribution to the spectrum is great enough to obscure a small effect.

Before the background correction was made, the spectrum for resonant capture exhibited a distinct line at  $62 \pm 1$  kev. However, as is observed in Fig. 8, this line is removed by the background correction. It is probable that the line results from the excitation of the first excited state of  $\text{I}^{127}$  by inelastic scattering of fast neutrons present as a contaminant in the beam of slow neutrons from the chopper. Perhaps some of the lines near 60 kev in the spectra of Fig. 5 are also caused by fast neutrons.

Having determined the isotopic origin of the lines for which delayed curves were obtained, we now attempt to fit the lines into a decay scheme. The first step is to establish that the 30-kev line, which might be thought to be from K x rays of iodine (27 kev), results mainly from a  $\gamma$  ray. This conclusion follows from the fact that the line is observed even when the NaI scintillator itself is used as the target. Under this condition an x ray could not give a pulse of about 30 kev because its pulse

would be added to that from the electron originating in the same process.

The close similarity of the half-lives measured for the lines at 30 and 137 kev, namely,  $8.8 \pm 1.0$  and  $8.0 \pm 0.6$  nsec, respectively, suggests that both of the lines originate from the decay of a single long-lived state. Two possible modes of decay are available for such a state but both modes are inconsistent with the data presented above. The emission of the 30- and 137-kev  $\gamma$  rays in cascade is excluded because, when the NaI scintillator is the target, the summing of pulses would cause the lines associated with the long lifetimes to have energies of 30 and 167 kev. As is shown in Fig. 5D, a delayed 167-kev line is not observed. The emission of the 30- and 137-kev gamma rays in parallel paths is rejected because there is no evidence for the delayed emission of the 107 kev of energy that would necessarily be in cascade with the 30-kev gamma rays. Thus it is concluded, in spite of the similarity of the two life-times, that the delayed  $\gamma$  rays at 30 and 137 kev result from the decay of different states. Since no other delayed lines are observed, these two states must be placed at energies of  $30 \pm 1$  and  $137 \pm 4$  kev. In view of the apparently high density of the low-lying states of  $\text{I}^{128}$  and the complexity of the pulse-height spectra, no attempt is made to fit the other observed lines into a decay scheme.

#### ACKNOWLEDGMENTS

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