Measure of K-Shell Internal Conversion Coefficients with a Coincidence Scintillation Spectrometer

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The K-shell internal conversion coefficients of gamma rays from Te^{123}, Sm^{153}, Hf^{181}, and Os^{191} have been measured using a coincidence scintillation spectrometer employing NaI and anthracene phosphors as detectors. In several cases an unambiguous classification of the transition as to multipole order and character is possible. The results indicate that the 159-kev gamma ray from Te^{123} is predominantly $M1$, the 70-kev gamma ray from Sm^{153} is a mixture of $E2+M1$, the 132-kev gamma ray from Hf^{181} is predominantly $E2$, the 135-kev gamma ray from Hf^{181} is predominantly $M1$, the 345-kev and 480-kev gamma rays from Hf^{181} are a mixture of either $E2+M1$ or $E1+M2$, and the 129-kev gamma ray from Os^{191} is a mixture of either $E2+M1$ or $E1+M2$.

I. INTRODUCTION

In many nuclear gamma transitions, a measurement of K-shell internal-conversion coefficients will permit an unambiguous assignment of multipole order and character of the transition. Recently, the importance of these measurements in conjunction with directional angular correlation measurements of nuclear gamma-ray cascades has become quite evident.[1] In some cases of a mixed multipole transition, a measured conversion coefficient will not distinguish between a parity-favored or a parity-unfavored transition, and the measurements must be supplemented by directional angular correlation measurements if possible.

For $Z \gtrsim 40$, many K-shell internal-conversion coefficients are easily measured with a scintillation spectrometer employing NaI phosphors. Many transitions have already been classified from measurements by this method.[2] For a simple decay scheme consisting of a single gamma ray, the K-shell internal-conversion coefficient is obtained from a pulse spectrum of the gamma radiation. However, in decay schemes involving gamma-ray cascades, a knowledge of the pulse spectrum of the gamma radiation from a particular transition is required. In many cases this spectrum can be obtained from a coincidence scintillation spectrometer. In this paper, a fast delayed coincidence scintillation spectrometer using NaI detectors is described. The application of coincidence scintillation spectrometry to the measurement of K-shell internal-conversion coefficients is discussed. Possible interferences in coincidence spectrometry are enumerated and discussed briefly. Experimental measurements of eight K-shell internal-conversion coefficients and a discussion of the results are presented.

II. APPARATUS

For the detection of the radiations, scintillation detectors consisting of Tl-activated sodium iodide crystals and an anthracene crystal mounted on RCA type 5819 photomultiplier tubes were used. The sodium iodide crystals were 1.5 in. in diameter by 1 in. thick, and the anthracene crystal was 1.5 in. in diameter by 0.20 in. thick. In most experiments the detectors were oriented at 180° with respect to the source which was located on the axis of the cylindrical crystals. The light shield assembly for the detectors was surrounded by 4 in. of Pb. To suppress the characteristic x-rays from the Pb shield by a factor of 10^3, the Pb shield was lined with a graded shield consisting of 0.090 in. of Cd and 0.015 in. of Cu.

Figure 1 is a block diagram of the delayed coincidence scintillation spectrometer which uses the fast-slow coincidence method. Although this apparatus has been in use for several years to measure the lifetimes[3] of short-lived metastable states of nuclei, it is perhaps appropriate to describe the fast-slow coincidence method, indicating its advantages in view of the advanced technique of scintillation spectrometry.

The integrated current pulse from each photomultiplier is fed both to a linear amplifier[4] and to a grid of a cathode follower. The coupling condenser between the cathode follower and the 200-ohm transmission

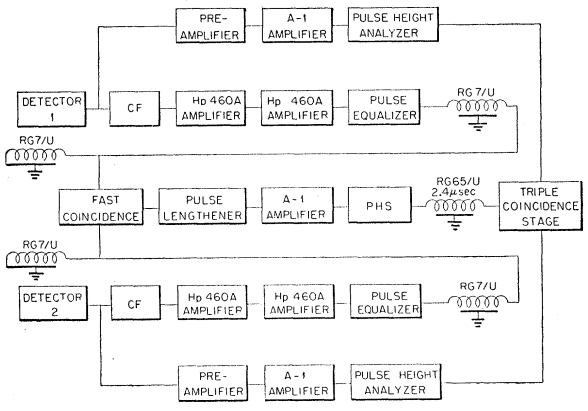

FIG. 1. Block diagram of the delayed coincidence scintillation spectrometer.

[1] F. R. Metzger and H. C. Amacher, Phys. Rev. **88**, 147 (1952); R. M. Steffen, Phys. Rev. **90**, 321 (1953).
[2] See for instance "The nuclear data, 1952 cumulation," Nuclear Science Abstracts **6**, No. 24B (1952).
[3] F. K. McGowan, Phys. Rev. **79**, 404 (1950).
[4] W. H. Jordan and P. R. Bell, Rev. Sci. Instr. **18**, 703 (1947).

line of the Hewlett-Packard 460A wide-band amplifiers is chosen to provide a time constant equal to the decay period of the phosphor. In this way the signal to be amplified is the current pulse which has been smoothed by some integration. This much integration assures one that the timing circuit is actuated by the arrival of the first n photoelectrons injected into the multiplier and is not actuated again by other groups of photoelectrons that may occur later in the same current pulse. More integration than this only makes the recovery time unnecessarily longer. After suitable amplification with two Hewlett-Packard 460A amplifiers in cascade, the pulses are equalized by limiting in a Western Electric 404A pentode. The equalization minimizes the error in determining the time of occurrence of the nuclear event which excites the phosphor crystal. The limited signal is led along a 100-ohm coaxial cable to a 100-ohm shorted coaxial cable clipper to fix the duration of the pulse. The pulses from either channel can be delayed by accurately known amounts by lengthening the appropriate signal cable. The clipped pulses are fed directly to a fast coincidence circuit consisting of a 1N21A diode. These unselected fast coincidence pulses are lengthened and amplified with a linear amplifier[4] equipped with a pulse-height selector. The minimum useful signal for the timing circuit corresponds to about $n=5$ photoelectrons injected into the multiplier. With NaI and anthracene as phosphors, the timing error for the arrival of the first five photoelectrons through the detectors is approximately $(4.5/E)\times10^{-7}$ sec and $(8/E)\times10^{-8}$ sec, respectively, where E is the energy in kev dissipated in the phosphor. These values of the timing error are, of course, for a single measurement. Since the phenomena being measured are repetitive, the timing error improves with the square root of the number of observations.

The two outer channels with the scintillation detectors represent the usual scintillation spectrometer. The differential pulse-height analyzer[5] selects all pulses whose amplitudes lie between E and $E+dE$. The output pulses of the analyzers are fed into a slow triple coincidence stage. The unselected fast coincidence pulses are delayed to match the delay introduced by the pulse-height analyzers before being fed to the slow coincidence stage. In this way only those coincidences are selected corresponding to signals from detectors 1 and 2 whose amplitudes after suitable amplification fall within the respective windows of the analyzers.

TABLE I. Ratio of the integral of the pulse spectrum in the full-energy peak to the integral of the total pulse spectrum.

Source	E_γ (kev)	Ratio of peak to total at 0.2 cm	Ratio of peak to total at 7.0 cm
Hg^{203}	279	0.769	···
Cr^{51}	320	0.690	···
Au^{198}	411	0.532	···
Sr^{85}	512	0.441	0.423
Cs^{137}	661	0.340	···
Cb^{95}	760	0.285	0.297
Mn^{54}	824	0.269	0.275
Zn^{65}	1114	0.219	0.215
K^{42}	1510	···	0.14
Pr^{144}	2180	···	0.087

[5] Francis, Bell, and Gundlach, Rev. Sci. Instr. **22**, 133 (1951).

Actually, the gain of the detectors is not a fixed quantity but is statistical in nature. This pulse-height spread would introduce an additional timing error if pulse-height selection of the pulses from the amplifier with a finite rise time preceded the timing circuit. The fast-slow coincidence method eliminates this timing error in that all pulse-height selection follows the timing circuit. Thus, the difficulties of fast pulse-height selection are avoided.

For gamma rays of a few hundred kev incident on a NaI phosphor, a time resolution of $2\tau_0=8.8\times10^{-9}$ sec without reducing the coincidence efficiency below unity has been realized. The trailing edge of a time-resolution curve for prompt events has a slope corresponding to a half-period of 5×10^{-10} sec. Under these conditions nuclear metastable states with $T_{\frac{1}{2}}>10^{-9}$ are measurable.

In some of the measurements of coincidence gamma-ray spectrums, where a small resolving time was not necessary, the wide-band amplifiers were not used. In this case the signals from the pulse-height selector of the linear amplifiers were fed to a pulse-shaping and coincidence circuit whose resolving time, $2\tau_0$, could be varied down to 2×10^{-7} sec for the fast coincidence portion of the coincidence spectrometer. The timing error introduced by the pulse-height selector preceding the fast coincidence stage was reduced to a minimum by adjusting the bias of the selector to trigger on all pulses just above the noise level of the amplifier.

In a typical experiment the pulse spectrum of the gamma radiation in coincidence with another gamma ray of the cascade was measured. The window of the fixed pulse-height analyzer was always operated to include only the full-energy-pulse spectrum peak of the gamma ray. The window width of the analyzer scanning the coincidence pulse spectrum was 12 pulse-height units out of 1500. Most of the data in these experiments were recorded automatically. The time for the collection of a fixed number of coincidence counts was printed on a paper tape by a printing timer which also advanced the fifteen-turn helipot of the pulse-height analyzer a fixed increment to the next energy. Unless otherwise specified, each point in a pulse spectrum contains 4096 counts.

III. DISCUSSION OF METHOD

The method of measuring the K-shell internal-conversion coefficients which has been used involves the measurement of the number of K x-rays and the number of gamma rays from the source per unit time. A measurement of the rate of emission of gamma radiation from a source requires a knowledge of the efficiency of the detector and a counting of the number of gamma rays registered by the detector. The latter is obtained from an integration of the entire differential pulse-

height spectrum, i.e., number of pulses observed per unit time per unit pulse height *versus* pulse height. For a monoenergetic gamma ray incident on the detector, the integration of the pulse spectrum is straightforward; but with a polyenergetic gamma-ray source the integration cannot be done easily. The full energy of a gamma ray is dissipated in the phosphor by photoelectric absorption and also by multiple processes, e.g., Compton recoil electrons and the eventual absorption of the Compton-scattered gamma ray. Since the pulse spectrum from a NaI scintillation detector for a gamma ray whose energy is totally absorbed in the phosphor appears to be Gaussian, it is desirable to know the ratio of the integral of the pulse spectrum in the full-energy peak to the integral of the total pulse spectrum as a function of the gamma-ray energy. In this way the total number of gamma rays detected by the scintillation counter can be obtained from an integration of the full-energy pulse spectrum of each gamma ray. The ratio of these two integrals has been measured by Bell, Heath, and Davis[6] for a NaI crystal of the size above and a source distance of 1 cm from the crystal face at eight different energies distributed between 0.141 and 1.114 Mev. To check the operation of the two NaI scintillation spectrometers used for the measurements below, the ratio of these two integrals (ratio of peak to total) was measured at several different gamma-ray energies. The results are tabulated in Table I. At a source distance of 0.2 cm from the front face of the crystal, the values of the ratio of peak to total are probably good to within ± 2 percent. At 7.0 cm the intensity of the pulse spectrum from degraded gamma radiation resulting from the scattering of the primary radiation by the surroundings becomes appreciable and must be removed from the gross pulse spectrum. As a result the values of the ratio of peak to total at 7.0 cm are probably uncertain by ± 5 percent with the exception of the last two entries, which are good only to within ± 10 percent.

Let: R_γ, R_x = ratio of peak to total for the gamma ray and the K x-ray, respectively; $N(E_0)_\gamma$, $N(E_0)_x$ = counting rate at the peak of the full-energy peak; a_γ, a_x = half-width of the full-energy peak at $1/e$ amplitude; ϵ_γ, ϵ_x = detection efficiency; A_γ, A_x = correction for absorption of gamma radiation in the beta shield and NaI crystal holder; w_K = fluorescent yield[7] of K x-rays. From these definitions the K-shell internal-conversion coefficient (ratio of the number of K-shell vacancies formed to the number of gamma-ray transitions per unit time) is

$$\alpha_{\exp}{}^K = \frac{N(E_0)_x n(E_0)_\gamma R_\gamma \epsilon_\gamma A_\gamma}{N(E_0)_\gamma n(E_0)_x R_x \epsilon_x w_K A_x}, \qquad (1)$$

where $n(E_0)_\gamma = 1/(a_\gamma \pi^{\frac{1}{2}})$ and $n(E_0)_x$ is similarly defined.

[6] Bell, Heath, and Davis, Physics Division Quarterly Progress Report, September 20, 1952, Oak Ridge National Laboratory Report ORNL-1415 (unpublished).

[7] Steffen, Huber, and Humbel, Helv. Phys. Acta. **22**, 167 (1949); Broyles, Thomas, and Haynes, Phys. Rev. **89**, 715 (1953).

TABLE II. The ratio of the number of K x-rays of iodine that leave the phosphor to the number of gamma rays detected by the phosphor.

Source	E(kev)	Experimental ratio	E(kev)	Computed ratio
Gd^{153}	41.6	0.238	35	0.268
			50	0.236
Tb^{160}	46.0	0.170	45	0.205
			50	0.180
Tm^{170}	52.5	0.149	55	0.156
			60	0.137
Lu^{177}	55.5	0.160	65	0.118
			70	0.102
Hf^{181}	57.5	0.130	75	0.090
			80	0.080
W^{187}	60.8	0.115	85	0.071
			90	0.063
Au^{198}	70.8	0.099	95	0.056
			100	0.049
Hg^{203}	73.1	0.103	110	0.040
			120	0.032
Bi^{207}	75.1	0.087	130	0.027
			140	0.021

For gamma radiation below 100 kev, the ratio of peak to total would be unity, provided the K x-ray of iodine did not escape from the detector. Actually the pulse spectrum of gamma radiation of this energy is made up of two peaks (the full-energy peak and the escape peak whose energy is the difference between the energy of the incident radiation and the K x-ray of iodine). The ratio of the number of K x-rays of iodine that leave the phosphor to the number of gamma rays detected by the phosphor was measured at eight different energies distributed between 41.6 and 75.1 kev. The results are shown in Table II. For comparison the computed ratio for a collimated beam of gamma radiation entering a semi-infinite crystal of NaI normal to its plane surface is also tabulated.

The detection efficiency has been computed as a function of the geometry and cross section of the NaI detector for gamma radiation by the Mathematics Panel at Oak Ridge National Laboratory. Computations were done for a right circular cylinder of height 1 in. and diameter 1.5 in. with the point source located on the axis of the cylindrical crystal at a distance h above the front face. It is assumed that the gamma radiation detected by the crystal is directly proportional to $(1-e^{-\tau x})$, where τ is the absorption cross section of the NaI crystal and x is the distance the radiation travels through the crystal. It is also assumed that there is no radiation loss between the source and the crystal. The detection efficiency with this arrangement is given by

$$\epsilon_\gamma(E) = \tfrac{1}{2} \int_0^\pi [1 - e^{-\tau(E)x(\alpha)}] \sin\alpha d\alpha.$$

The efficiency $\epsilon_\gamma(E)$ has been evaluated at each distance $h = 0$, 0.2, 0.3, 0.5, 0.7, 1.0, 1.5, 2.0, 3.0, 5.0, 7.0, 10.0, 20.0, and 50.0 cm for 14 values of τ distributed

between 600 and 0.123 cm^{-1}.[8] The absorption cross section of NaI for gamma radiation as a function of gamma-ray energy was obtained from the tables compiled by White[9] and from transmission measurements by Bell *et al.*[10]

So far nothing has been mentioned about possible interferences that arise in coincidence scintillation spectrometry. One must bear in mind that there are a number of possible false coincidences which will lead to erroneous results unless they are either eliminated from the measurements or are properly removed from the gross coincidence spectrums. A few of the difficulties encountered in the measurements discussed in this paper are:

(1) Scattering of gamma radiation from one detector to the other.

(2) Scattering from the shield and surroundings into the detector.

(3) Sum pulses either by simultaneous detection of several coincidence radiations or by random detection of several radiations.

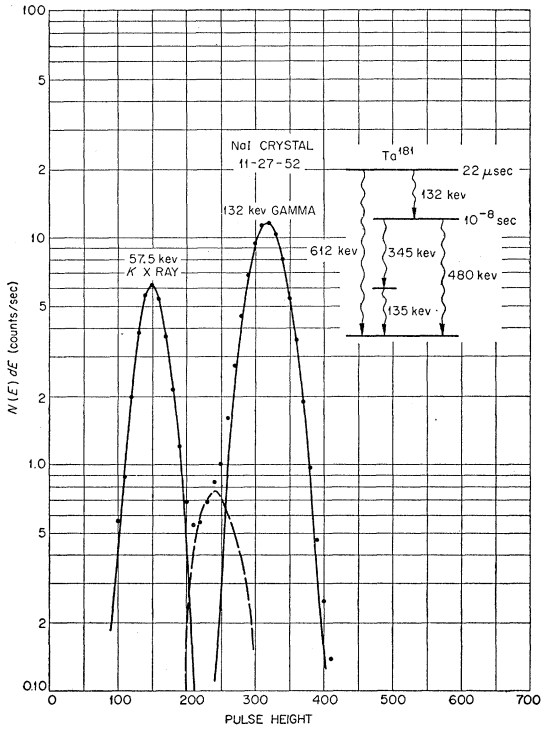

FIG. 2. Differential pulse-height spectrum of the gamma radiation announcing the formation of the 10^{-8} sec state of Ta^{181} in delayed coincidence with the 480-kev gamma ray. Each point in the spectrum contains about 10^4 coincidence counts. The inset is the Ta^{181} decay scheme.

[8] Tables of the efficiencies are available for distribution from either P. R. Bell or the writer.

[9] G. R. White, National Bureau of Standards Report NBS-1003, 1952 (unpublished).

[10] Bell, Hughes, Davis, Jordan, and Randall, Physics Division Quarterly Progress Report, September 20, 1952, Oak Ridge National Laboratory Report ORNL-1415 (unpublished).

(4) Iodine K x-rays escaping from one detector into the other detector.

(5) Characteristic x-rays from the shield.

(6) Anisotropy effects from the directional angular correlation of the gamma rays.

These difficulties are discussed in more detail under the section on experimental measurements.

IV. EXPERIMENTAL MEASUREMENTS AND DISCUSSION OF RESULTS

A. Hf^{181}

The radiations from the decay of Hf^{181} have been studied by many investigators.[11] In spite of the numerous activities produced by neutron-irradiated Hf, the general features of the decay scheme of Hf^{181} given originally by Chu and Wiedenbeck[12] are probably correct.[13] This decay scheme is shown in the inset of Fig. 2.

Sources of Hf^{181} were prepared from a sample of HfO_2 irradiated with pile neutrons for four weeks. The sample of HfO_2 was enriched in Hf^{180} (93.96 percent).[14] With this enrichment the intensity of the 342-kev gamma ray of Hf^{175} in the source relative to the 345-kev gamma ray

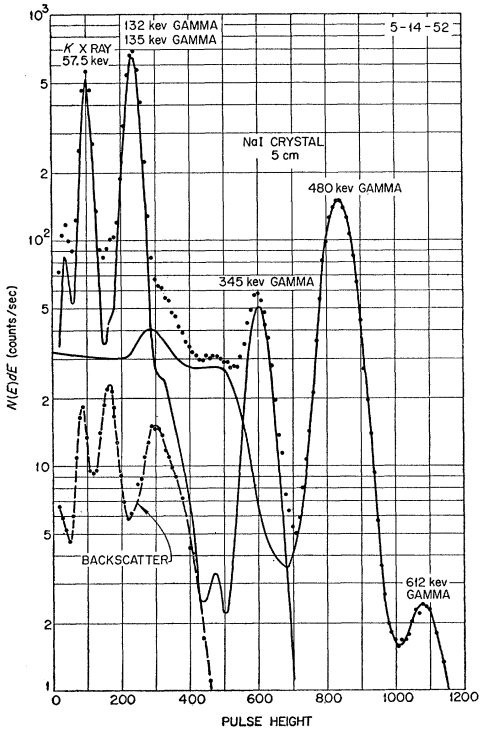

FIG. 3. A differential pulse-height spectrum of the gamma radiation from $Hf^{181} \rightarrow Ta^{181}$.

[11] *Nuclear Data*, National Bureau of Standards Circular 499 (U. S. Government Printing Office, Washington, D. C., 1950) and Supplements 1, 2, and 3.

[12] K. Chu and M. Wiedenbeck, Phys. Rev. **75**, 226 (1949).

[13] Arne Hedgran and Sigvard Thulin, Phys. Rev. **81**, 1072 (1951).

[14] Obtained from the Isotopes Division of the Oak Ridge National Laboratory.

TABLE III. Experimental and theoretical internal-conversion coefficients.

Nucleus	E_γ (kev)	$\alpha_{exp}{}^K$	Theoretical coefficients $\alpha_l{}^K$ and $\beta_l{}^K$	Classification
$_{52}Te^{123}$	159	0.19±0.02	$\alpha_2=0.260$, $\beta_1=0.175$	$M1$
$_{63}Eu^{153}$	70	3.8 ±0.2	$\alpha_1=0.63$, $\alpha_2=2.9$, $\beta_1=5.7$	$E2+M1$
$_{63}Eu^{153}$	102	1.14±0.20	$\alpha_1=0.25$, $\alpha_2=1.1$, $\beta_1=1.6$	···
$_{73}Ta^{181}$	132	0.48±0.02	$\alpha_1=0.15$, $\alpha_2=0.495$, $\beta_1=1.94$	$E2$
$_{73}Ta^{181}$	135	1.88±0.20	$\alpha_1=0.15$, $\alpha_2=0.495$, $\beta_1=1.94$	$M1$
$_{73}Ta^{181}$	345	8×10^{-2}	$\alpha_1=1.35\times10^{-2}$, $\alpha_2=4.03\times10^{-2}$, $\alpha_3=11.2\times10^{-2}$ $\beta_1=14.2\times10^{-2}$, $\beta_2=46.0\times10^{-2}$	···
$_{73}Ta^{181}$	480	$(3.4\pm0.4)\times10^{-2}$	$\alpha_1=0.64\times10^{-2}$, $\alpha_2=1.74\times10^{-2}$, $\alpha_3=4.40\times10^{-2}$ $\beta_1=6.00\times10^{-2}$, $\beta_2=16.4\times10^{-2}$, $\beta_3=40.2\times10^{-2}$	$E2+M1$ or $E1+M2$
$_{77}Ir^{191}$	129	2.07±0.10	$\alpha_2=0.518$, $\alpha_3=1.27$, $\beta_1=3.10$	$M1+E2$

of Hf^{181} is about 4 percent. With sources of normal Hf, the intensity of the 342-kev gamma ray is equal to the 345-kev gamma-ray intensity three weeks after the end of the irradiation.

A differential pulse-height spectrum of the gamma radiation from $Hf^{181}\rightarrow Ta^{181}$ is shown in Fig. 3. The dashed curve is the pulse spectrum of degraded gamma radiation resulting from Compton scattering of the primary radiation by the surroundings. The degraded spectrum was obtained by inserting a cone of Pb between the source and detector to shadow the phosphor from the direct radiation. This degraded spectrum is not the full intensity of the degraded radiation incident on the phosphor because the cone also shadows the surroundings beyond the back face of the phosphor, but it does indicate the general shape and magnitude. The full-energy peak at 612 kev is not the true intensity of the 612-kev crossover transition but is partly the result of simultaneous detection of the 132- and 480-kev gamma rays. For the purpose of obtaining the true intensity of the crossover transition, the source distance is increased until the intensity of the full-energy peak decreases proportionally with ϵ_{612}, whereas the sum pulse intensity decreases with the product $\epsilon_{132}\epsilon_{480}$. From a measurement of the pulse spectrum with the source at 30 cm, the intensity of the 612-kev gamma radiation relative to the 480-kev gamma radiation is 0.8 percent.

For analysis of a pulse spectrum into its components, the following procedure has been used. A set of Gaussian templets made up on 3-cy semilog paper is used to fit the full-energy pulse-spectrum peaks visually. A Compton recoil electron distribution plus backscatter is constructed from the pulse spectrum of a monoenergetic gamma-ray source with energy as near as possible to the energy of the gamma-ray whose pulse-spectrum shape one wants to know. The intensity of the Compton distribution is adjusted to fit the measured peak-to-total ratio taken from a curve of the data in Table I. The pulse spectrum of the most energetic gamma ray is peeled off from the gross spectrum. The process is repeated for the next gamma ray down in energy, and so on for the rest of the gamma rays making up the pulse spectrum. From the pulse spectrum of Fig. 3, the intensity of the 345-kev gamma radiation relative to 480-kev gamma radiation is 15.7 percent.

Figure 2 is the differential pulse spectrum of the gamma radiation announcing the formation of the 10^{-8}-sec state of Ta^{181} in delayed coincidence with the 480-kev gamma ray at a time delay 1.2×10^{-8} sec which is sufficient to resolve simultaneous events. NaI detectors were employed in both channels and the resolving time $2\tau_0$ was 1.32×10^{-8} sec. In addition to the pulse spectrum of the 132-kev gamma ray and K x-rays from the internal conversion of this gamma ray, there is a composite peak at 240 pulse-height units because of the backscatter and escape peak of the 132-kev gamma ray. The K-shell internal-conversion coefficient obtained from an analysis of this pulse spectrum is tabulated, together with the theoretical internal conversion coefficients[15] in Table III. The transition is classified as predominantly $E2$ radiation. Previous measurements[16] of α^K (132 kev) using a coincidence scintillation spectrometer with $2\tau_0=3.8\times10^{-7}$ sec were always somewhat larger because of the sum pulse resulting from 132+345-kev gamma rays which could not be distinguished from the full-energy pulse of the 480-kev gamma ray.

The K-shell internal-conversion coefficient of the 480-kev gamma ray was obtained from intensity measurements of the K-shell internal-conversion electrons and the gamma ray with an anthracene and NaI scintillation spectrometer, respectively. In one set of experiments the intensities were measured from the singles pulse spectrum as shown in Fig. 4. The conversion-electron pulse spectrum above the endpoint of the beta spectrum is resolved into two Gaussian components ($L+M$ conversion electrons and K conversion electrons). The $K/(L+M)$ ratio of 4.0±0.6 is obtained directly from an integration of these two components. Only the full-energy pulse spectrum peak of the 480-kev gamma ray is shown in Fig. 4. In another set of experiments these intensities were obtained from the pulse spectra in coincidence with the 132-kev gamma ray as shown in Fig. 5. The agreement between the two sets of measurements was good. To check the method of measurement the K-shell internal-conversion coefficient

[15] Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. **83**, 79 (1951).

[16] F. K. McGowan, Physics Division Quarterly Progress Report, December 20, 1951, Oak Ridge National Laboratory Report ORNL-1278 (unpublished).

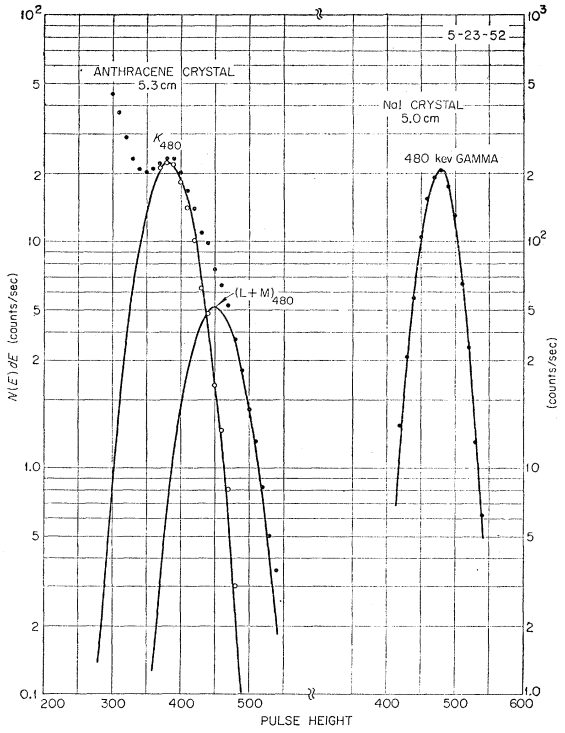

FIG. 4. Differential pulse-height spectrum of the internal conversion electrons and gamma radiation from the 480-kev transition of Ta^{181}.

of the 661-kev gamma transition of Ba^{137m} was measured. The result agreed to within 6 percent of the accepted $M4$ assignment for the transition. In all experiments involving intensity measurements of conversion electrons, the air between the source and detector was displaced with H_2 gas.

A similar set of measurements was done to determine α^K (345 kev) but with considerably less precision. The results are tabulated in Table III. From the conversion coefficients, the 480-kev transition may be interpreted as either $E2+M1$ with $E2$ to $M1$ intensity ratio of 1.57 or $E1+M2$ with $M2$ to $E1$ intensity ratio of 0.21 or $E2+M3$ with $M3$ to $E2$ intensity ratio of 0.046. A classification $E2+M3$ would require the $M3$ transition probability to be 1.6×10^4 times greater than estimates from the independent-particle model[17] and therefore seems rather unlikely. The theoretical estimates represent upper limits to the transition probabilities. Results from extensive directional angular correlation measurements[18] of the various gamma-ray cascades of Ta^{181} require the interpretation that 480-kev transition is a mixed multipole. A similar interpretation is probable for the 345-kev transition.

[17] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952).

[18] F. K. McGowan, Physics Division Quarterly Progress Reports, Oak Ridge National Laboratory Reports ORNL-1365, 1415, and 1496. These results will be discussed in a subsequent paper in the near future.

A differential pulse-height spectrum of all delayed radiation from the 10^{-8}-sec metastable state in delayed coincidence with the 132-kev gamma ray is shown in Fig. 6. After the pulse-spectrum shapes for the 480- and 345-kev gamma rays are peeled off from the total spectrum of delayed radiation, there is left the pulse spectrum of the 135-kev gamma ray and K x-ray. The K x-rays, of course, result from internal conversion of all three gamma rays (480, 345, and 135 kev). Since the K-shell internal-conversion coefficients of the 345- and 480-kev gamma rays are known from the measurements discussed above, α^K (135 kev) is obtained from the analysis of this pulse spectrum. The result is tabulated in Table III, and the transition is classified as predominantly $M1$ radiation.

Difficulties 1, 3, and 4 in Sec. III are not, in general, encountered in the delayed coincidence spectrum measurements of Hf^{181} aside from sum pulses by random detection of several radiations. In the spectrum of delayed gamma radiation of Fig. 6 the sum pulses due to simul taneous detection of 345- and 135-kev gamma rays do occur, and in the analysis of this spectrum a correction for this effect has been included. The anisotropy effects are also very small and may be neglected. For instance, the largest anisotropy observed in any one of the cascades for a solid source of $Hf^{181}(OH)_4$ is 10 percent.[18] For the angular resolution used in the measurements

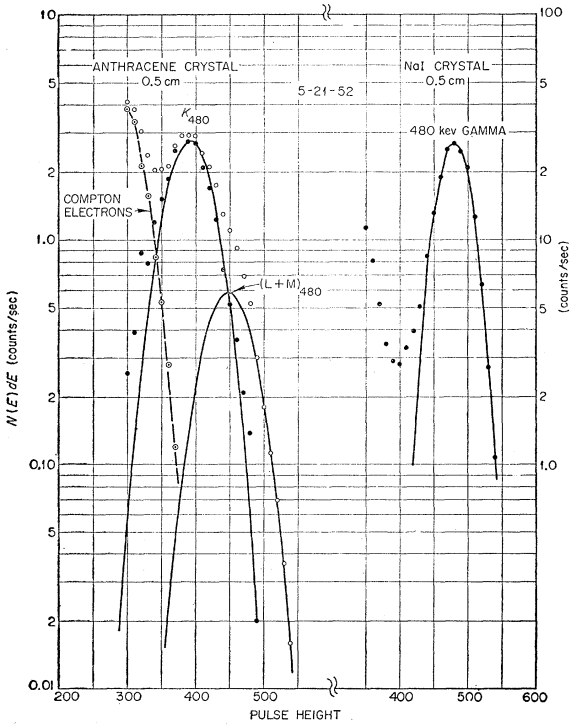

FIG. 5. Differential pulse-height spectrum of the internal-conversion electrons and gamma radiation from the 480-kev transition of Ta^{181} in coincidence with the 132-kev gamma ray. Each point in the spectrum contains 1024 coincidence counts.

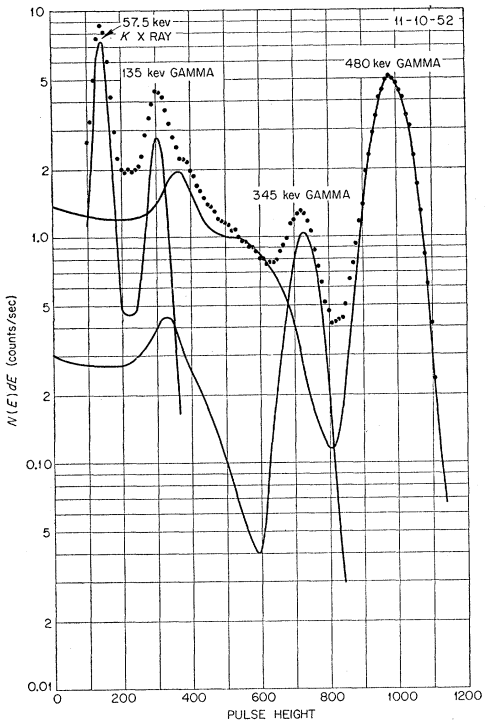

FIG. 6. Differential pulse-height spectrum of all delayed radiation from the 10^{-8}-sec metastable state of Ta^{181} in delayed coincidence with the 132-kev gamma ray. Each point in the spectrum contains 4096 coincidence counts.

above, any corrections to the gamma-ray intensities are less than 0.6 percent.

While these K-shell internal-conversion coefficients were being measured, Fan[19] reported values for the conversion coefficients obtained from electron intensity measurements. Fan concluded that all the transitions were pure multipoles, a conclusion not in accord with measurements above. Also, in those cases for which we disagree, the results[18] from the directional angular correlation measurements require that the transitions be a mixture of two different multipoles.

B. Sm^{153}

Although much work[20] on the decay of Sm^{153} has been done, it is apparent that the data are not all consistent. At least some of the beta emission of Sm^{153} leads to a metastable state in Eu^{153} which decays with a $T_{\frac{1}{2}}=3\times10^{-9}$ sec[21] by the emission of two transitions, 70 kev and 102 kev, in cascade. For the measurements and results discussed below, a knowledge of either the correct decay scheme or order of emission of the 70-kev and 102-kev transitions is not needed.*

Sources of Sm^{153} were prepared from samples of Sm_2O_3 irradiated with pile neutrons. The samples of Sm_2O_3 were purified by Boyd and Harris of the Chemistry Division of Oak Ridge National Laboratory. A differential pulse-height spectrum of the gamma radiation in coincidence with the 102-kev gamma ray is shown in Fig. 7. The data were taken with a coincidence scintillation spectrometer with a resolving time $2\tau_0=3.8\times10^{-7}$ sec with NaI detectors being used in both channels. The K-shell internal-conversion coefficient of the 70-kev transition obtained from an analysis of this pulse spectrum is tabulated in Table III. The theoretical K-shell conversion coefficients for $E_\gamma=70$ and 102 kev are extrapolated values. The method[22] of extrapolation has been described previously. The transition is classified $E2+M1$ with $E2$ to $M1$ intensity ratio of 2.0. A classification $E1+M2$ with $M2$ to $E1$ intensity ratio of 0.074 seems rather unlikely because the observed $M2$ transition probability is at least 5×10^2 times greater than estimates from the independent-particle model.[17]

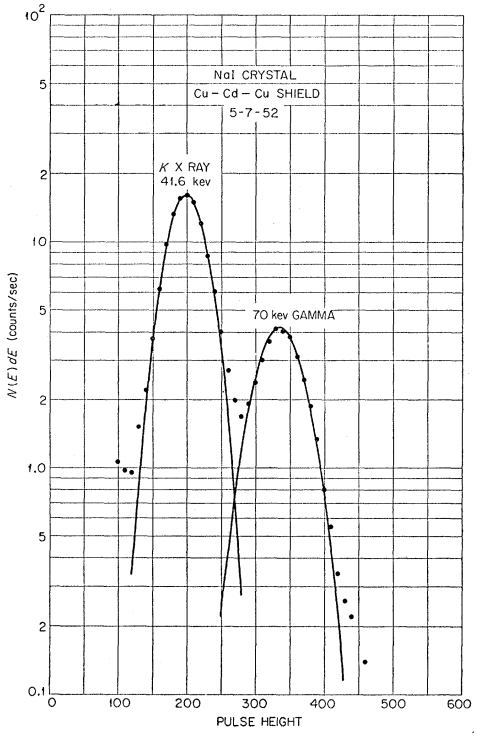

FIG. 7. Differential pulse-height spectrum of the gamma radiation in coincidence with the 102-kev gamma from $Sm^{153}\rightarrow Eu^{153}$. Each point in the spectrum contains 2048 coincidence counts.

[19] C. Y. Fan, Phys. Rev. **87**, 252 (1952).

[20] *Nuclear Data*, National Bureau of Standards Circular 499 (U. S. Government Printing Office, Washington, D. C., 1950) and Supplements 1 and 2. New Nuclear Data 1952 Cumulation, Nuclear Science Abstracts **6**, No. 24B (1952).

[21] F. K. McGowan, Phys. Rev. **80**, 482 (1950).

* *Note added in proof.*—Recent measurements with the delayed coincidence scintillation spectrometer have shown conclusively that the 70-kev transition precedes the 102-kev isomeric transition of 3.4×10^{-9} sec half-life. Also, the 102-kev transition following the ϵ-capture decay of Gd^{153} decays with $T_{1/2}=3.4\times10^{-9}$ sec. In addition, the ϵ-capture decay of Gd^{153} leads to another excited state in Eu^{153} within a few kev of the 102-kev level. The gamma ray from the decay of this state is in prompt coincidence ($T_{1/2}<10^{-9}$ sec) with the K x-rays from ϵ capture and is 1.4 times as intense as the 102-kev gamma ray.

[22] F. K. McGowan, Phys. Rev. **85**, 151 (1952).

Coincidence spectra of gamma radiation in coincidence with a gamma ray of higher energy are, in general, free of difficulty (1) (see Sec. III) and are much easier to analyze. For the reverse situation, coincidence spectra of gamma radiation in coincidence with a gamma ray of intermediate energy, difficulty (1) is very troublesome unless the NaI phosphors are arranged so that they do not see each other for gamma radiation. Pulse spectra of the gamma radiation in coincidence (a) with the 70-kev gamma ray and (b) with the K x-ray from internal conversion of the 70-kev transition were taken in an attempt to measure α^K (102 kev). The data were difficult to analyze. Unfortunately, in this case, the energies of the K x-rays and gamma rays are such that the escape peaks of the two gamma rays coincide in energy with the 70-kev gamma ray and K x-ray full-energy pulse-spectrum peaks, respectively. This situation gives rise to several sources of unwanted coincidences in the coincidence spectrum. In addition, the iodine K x-rays escaping from one detector to the other were troublesome. For the purpose of suppressing these, the source was mounted in a small aperture of a graded shield of 0.015 in. of Cu—0.010 in. of Cd—0.015 in. of Cu between the detectors. The unwanted coincidences in the spectrums taken under the conditions (a) and (b) were removed in the analysis, and good agreement was obtained between the two measurements for the K-shell internal-conversion coefficient ($\alpha^K=1.3$).

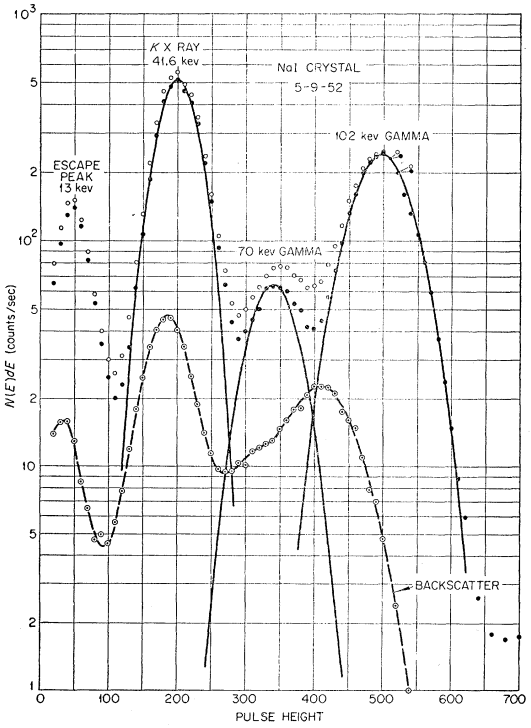

FIG. 8. Differential pulse-height spectrum of the low-energy gamma radiation from $Sm^{153}\rightarrow Eu^{153}$.

Since the value of α^K (70 kev) is known from the measurement above, a measurement of α^K (102 kev) with fewer uncertainties may be obtained from the singles pulse spectrum of the gamma radiation from Sm^{153}. Only the low-energy portion of the pulse spectrum is shown in Fig. 8. The value of α^K (102 kev) is tabulated in Table III, but the transition is not classified as to multipole order or character. Both the experimental value and the values of the extrapolated coefficients are too uncertain.

Preliminary measurements of the directional angular correlation of the 70- and 102-kev cascade indicate that the distribution is isotropic to within ± 3 percent.

C. Te^{123}

The 104-day Te^{123} isomeric state has been identified as $h_{11/2}$[23] which decays by the emission of an 88-kev $M4$ transition and a 159-kev transition in cascade. Katz, Hill, and Goldhaber have classified the 159-kev transition as $M1$ on the basis of its internal-conversion coefficient and K/L ratio.[24] The K-shell internal-conversion coefficient was obtained from electron intensity measurements by Katz *et al.* and based on the assumption of complete internal conversion of the 88-kev transition. The large uncertainty in their result would not exclude an admixture of $E2$ in the 159-kev transition.

A source of Te^{123} was prepared from a sample of Te enriched in Te^{122} (81.7 percent)[25] irradiated with neutrons. For the purpose of obtaining the spectrum of the gamma radiation associated with the 159-kev transition, it is necessary, of course, to measure the pulse spectrum in coincidence with only the L and M internal-conversion electrons from the $M4$ transition. An anthracene scintillation spectrometer has sufficient energy resolution to resolve the K internal-conversion electrons from the L and M internal-conversion electrons. The pulse spectrum of the gamma radiation from the 159-kev transition in coincidence with L and M internal-conversion electrons of the 88-kev transition is shown in Fig. 9. The peak at 320 pulse-height units is the maximum of the Compton recoil electron spectrum, and that at 580 pulse-height units is the backscatter peak due to large-angle Compton scattering of the 159-kev gamma ray from the surroundings. The K-shell internal-conversion coefficient obtained from an integration of this pulse spectrum of the K x-ray and the gamma ray is tabulated in Table III. The data indicate that the transition is predominantly $M1$ but do not exclude an admixture of $E2$. A sensitive test for this admixture would be a measurement of the directional angular correlation of the gamma-ray cascade. For instance, an intensity ratio of $E2$ to $M1$ of 1 percent changes the anisotropy [-0.21 for the sequence $11/2(M4)3/2(M1)1/2$] by ± 30 percent, depending on the sign of the amplitude

[23] R. D. Hill, Phys. Rev. **76**, 333 (1949).
[24] Katz, Hill, and Goldhaber, Phys. Rev. **78**, 9 (1950).
[25] Obtained from the Isotopes Division of Oak Ridge National Laboratory.

ratio of the multipoles in the mixture. Since the $M4$ transition is almost totally converted, the only angular correlation measurement possible is the conversion electron-gamma. For this the predicted anisotropy[26] is a few percent larger than the gamma-gamma anisotropy. An attempt was made to measure the directional angular correlation between the K-shell internal-conversion electron of the 88-kev transition and the 159-kev gamma ray. With a source of Te^{123} several mg/cm^2 thick, the angular distribution was isotropic to within ± 2 percent. The nearly complete attenuation of the angular correlation is attributed to the multiple scatter-

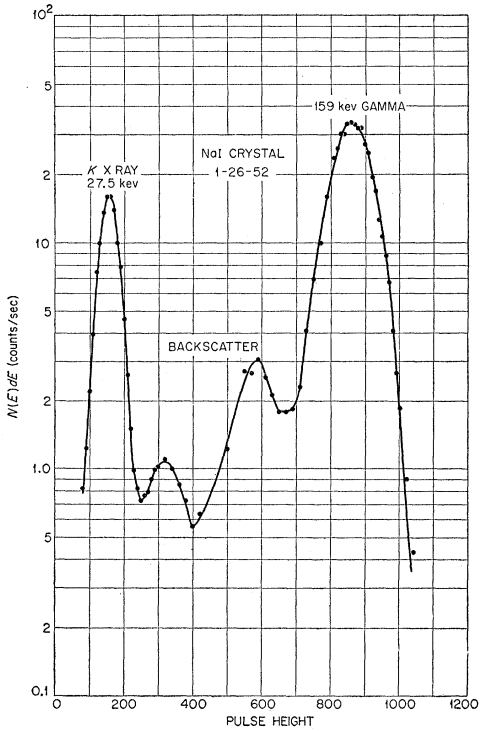

FIG. 9. Differential pulse-height spectrum of the gamma radiation from Te^{123} in coincidence with the L and M internal-conversion electrons of the 88-kev transition. Each point in the spectrum contains 512 coincidence counts except the points near the full-energy peaks of 27.5 kev and 159 kev which contain 4096 coincidence counts.

ing of the conversion electrons in the source and source backing.

D. Os^{191}

Os^{191} (15-day) is known to decay by beta emission followed by gamma rays of 42 and 129 kev.[11] The following values of the K-shell internal-conversion coefficient have been reported: 0.5 by Kondaiah,[27] 3.2 by Johansson,[28] 1.36 by Swan and Hill,[29] and 2.0 by

[26] Rose, Biedenharn, and Arfken, Phys. Rev. **85**, 5 (1952).
[27] E. Kondaiah, Arkiv. Fysik **3**, 47 (1951).
[28] S. A. E. Johansson, Arkiv. Fysik **3**, 533 (1952).
[29] J. B. Swan and R. D. Hill, Phys. Rev. **88**, 831 (1952).

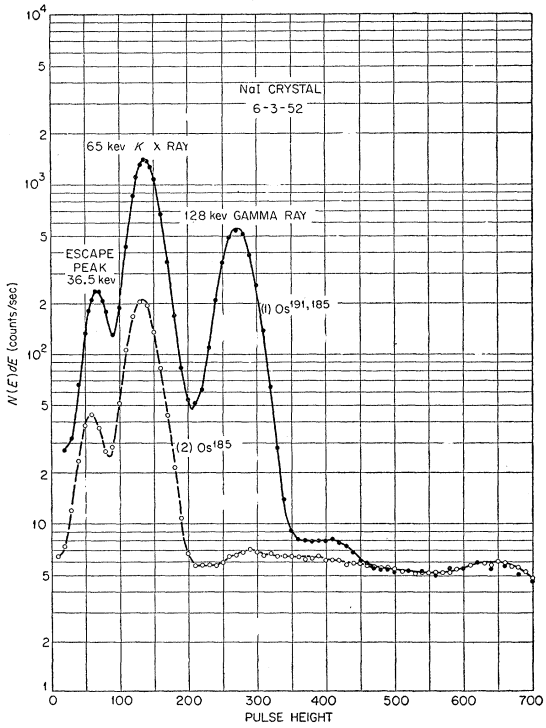

FIG. 10. Differential pulse-height spectrum of the gamma radiation from $Os^{191,185}$ and Os^{185}.

Hill and Mihelich.[30] Kondaiah also observed coincidences between the beta rays and conversion electrons. Earlier measurements[31] with a coincidence scintillation spectrometer employing anthracene detectors indicated that the conversion electrons from the 128-kev transition were not in coincidence with the beta rays.

The measurements have been repeated with a NaI and anthracene coincidence scintillation spectrometer. No coincidences between the beta rays and the 128-kev gamma ray have been observed. A measurement of the K-shell internal-conversion coefficient was obtained from the singles pulse spectrum of the gamma radiation from Os^{191}. The source was a sample of osmium tetraoxide irradiated with pile neutrons for two weeks. Curve 1 of Fig. 10 is the differential pulse-height spectrum of the gamma radiation from $Os^{191,185}$. Curve 2 is the differential pulse-height spectrum of the gamma radiation from Os^{185} (97-day) which is an ϵ-capture activity. The Os^{185} spectrum was normalized to the full-energy peaks of the 0.65- and 0.88-Mev γ-rays of Os^{185} (not shown) of curve 1. The K-shell internal conversion coefficient for the 129-kev transition obtained from the data shown in Fig. 10 is tabulated in Table III. The transition has been classified $M1+E2$ with the intensity ratio of $E2$ to $M1$ equal to 0.66. However, the

[30] R. D. Hill and J. W. Mihelich, Phys. Rev. **89**, 323 (1953).
[31] F. K. McGowan, Oak Ridge National Laboratory Report ORNL-952, 1951 (unpublished), p. 49.

results presented here do not exclude the possible classification $E1+M2$.

V. CONCLUSIONS

Of the eight K-shell internal-conversion coefficients measured, only three of the transitions may be classified as predominantly pure multipole radiation. In the other cases the transitions must be classified as mixed multipole radiation. Unfortunately, the values of the conversion coefficients for some of these cases do not distinguish between mixtures involving parity-favored and parity-unfavored transitions. The directional angular correlation measurements,[18] in combination with the conversion coefficients, will probably remove the ambiguity in the classification of the 345-kev and 480-kev gamma rays of Ta^{181}.

ACKNOWLEDGMENTS

It is a pleasure for the author to acknowledge his appreciation to P. R. Bell and G. G. Kelley for constant advice on the development of the apparatus and for numerous stimulating discussions concerning the measurements. In particular, their development of the scintillation spectrometer for measuring beta- and gamma-ray spectra has made it possible to extend the method of delayed coincidences to measure time intervals in the millimicrosecond region.

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The Nuclear Magnetic Moments of K^{41}, Y^{87}, Ag^{107}, and Ag^{109}

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(Received September 1, 1953)

The gyromagnetic ratios of K^{41}, Y^{89}, Ag^{107}, and Ag^{109} have been measured by nuclear induction technique. With the known spin values the magnetic moments without diamagnetic corrections are:

$$\mu_{41}=-0.21453\pm0.00003 \text{ nm},$$
$$\mu_{89}=-0.136825\pm0.000004 \text{ nm},$$
$$\mu_{107}=-0.113014\pm0.000004 \text{ nm},$$
$$\mu_{109}=-0.129924\pm0.000004 \text{ nm}.$$

In metallic silver a paramagnetic shift of 0.53 percent is observed.

THE nuclear magnetic moments of K^{41}, Y^{87}, Ag^{107}, and Ag^{109} have been measured with a nuclear induction type spectrometer of greatly increased sensitivity of a design similar to one described by Weaver.[1] With the exception of K^{41}, whose gyromagnetic ratio was determined relative to K^{39}, the resonance frequencies were measured at a fixed magnetic field of about 9000 gauss in terms of the proton resonance frequency in a sample of 0.1-molar $MnSO_4$, which was located immediately adjacent to the sample containing the unknown substance.

The results quoted below were obtained with the sufficiently well known values of the nuclear spins from hfs and the diamagnetically uncorrected value of the proton magnetic moment of Sommer, Thomas, and Hipple[2] $\mu_P=2.79268\pm0.00006$ nm. No diamagnetic corrections have been applied. In every case the sign of the magnetic moment was determined by comparison with the signal of a known moment.

Resonances of the two potassium isotopes 39 and 41 were obtained in a 15-molar aqueous solution of KCO_2H. Both isotopes are known to have spin $\frac{3}{2}$. For K^{39} the magnetic moment measured relative to proton was found to be

$$\mu_{39}=+0.390873\pm0.000013 \text{ nm},$$

in agreement with the value of Collins[3]: $\mu_{39}=+0.39094\pm0.00007$. The signal of K^{41} (abundance 6.9 percent) appeared with a signal-to-noise ratio of about 3:1. The ratio of the frequencies of the two isotopes is

$$\nu_{41}/\nu_{39}=0.54886\pm0.00008,$$

in agreement with the value 0.54891 ± 0.00005, as found from atomic beam measurements by Ochs, Logan, and Kusch.[4] The nuclear spin is $I=\frac{3}{2}$, and with the above value of μ_{39} one obtains

$$\mu_{41}=0.21453\pm0.00003 \text{ nm}.$$

Signals of the only stable isotope Y^{89} were obtained in a pure 3.3 molar aqueous solution of $Y(NO_3)_3$ with a signal-to-noise ratio of 20:1. The ratio of Yttrium to proton resonance frequency is

$$\nu_{89}/\nu_P=0.048994\pm0.000001.$$

[1] H. E. Weaver, Phys. Rev. **89**, 923 (1953).
[2] Sommer, Thomas, and Hipple, Phys. Rev. **82**, 697 (1951).
[3] T. L. Collins, Phys. Rev. **80**, 103 (1950).
[4] Ochs, Logan, and Kusch, Phys. Rev. **78**, 184 (1950).