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Internal-Conversion and γ-γ Directional-Correlation Studies in the Decay of ^{103}Ru†

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Internal-conversion coefficients, γ-ray intensities of weak γ rays, and two γ-γ directional correlations in the transitions of ^{103}Rh, after the β^- decay of ^{103}Ru, were measured. The conversion coefficients are: $\alpha_K(53) = 2.47 \pm 0.14$, $\alpha_K(444) = (6.4 \pm 2.7) \times 10^{-3}$, $\alpha_K(497) = (4.4 \pm 0.4) \times 10^{-3}$, $\alpha_K(557) = (3.7 \pm 0.7) \times 10^{-3}$, $\alpha_K(610) = (2.7 \pm 0.3) \times 10^{-3}$, and $K/L + M + (497) = 8.43 \pm 0.40$. The γ-γ directional correlations of the 557-53 and 444-53-keV cascades were measured with a 33-cm^3 Ge(Li) detector and a 2×2-in. NaI(Tl) detector on an automatic cycling correlation apparatus. The results are $A_2(557\text{-}53) = -0.130 \pm 0.008$ and $A_2(444\text{-}53) = -0.23 \pm 0.02$. The data imply that the 444-keV transition is pure $E2$ while the 53-keV transition is mixed $M1+E2$ with $\delta(53) = -0.13 \pm 0.03$. The data allow unambiguous spin and parity assignments of $\frac{9}{2}^+$, $\frac{5}{2}^+$, and $\frac{7}{2}^+$ for the 93-, 537-, and 650-keV levels, respectively. The above data imply that the 557-keV transition is a multipole mixed transition with $\delta(557) = -0.32 \pm 0.03$.

I. INTRODUCTION

There have been many investigations of the level scheme of ^{103}Rh. Unfortunately, these investigations have produced some conflicting results, and consequently definite spin and parity assignments could not be made for three of the six levels populated in the decay of ^{103}Ru. Excellent reviews of the earlier work can be found in two recent articles.[1,2] The primary purpose of the research presented in this paper was to measure the 444-53 and 557-53-keV γ-γ directional correlations accurately enough to uniquely fix the spins of the 93-, 537-, and 650-keV levels. In addition, remeasurements were made of the internal-conversion coefficients which play a role in the assignments of J^π for these levels.

The spin of the ground state was first measured as $\frac{1}{2}$ by Kuhn and Woodgate[3] using atomic-beam methods. Because this state appears to have the properties of a single unpaired proton in a $2p_{1/2}$ state, a spin and parity assignment of $\frac{1}{2}^-$ was made.

The first excited state of ^{103}Rh is a 57-min isomeric level at 40 keV. There is much evidence supporting an $E3$ multipolarity assignment for the 40-keV transition to the ground state. The original multipolarity assignment was made by Goldhaber and Sunyar[4] on the grounds of the empirical relationship between K/L internal-conversion ratios and isomeric half-lives. Coulomb-excitation studies of Jones and Phillips[5] as well as the L-subshell measurements of Manthuruthil, Hennecke, and Cothern[1] and internal-conversion-coefficient measurements of Lepri and Lyon[6] and those of Nieschmidt and Pearson,[7] have all confirmed this multipolarity. The spin and parity of the 40-keV level is thus $\frac{7}{2}^+$. Manthuruthil, Hennecke, and Cothern[1] conclude, on the basis of their internal-conversion studies, that the level at 93 keV should be assigned a spin and parity of $\frac{7}{2}^+$. However, Zoller *et al.*[2] argue in favor of a $\frac{9}{2}^+$ assignment on the basis of their γ-γ directional-correlation and β-decay studies. The spins and parities of the levels at 295 and 358 keV have been established by Edwards and Boehm[8] from their measurements of angular distributions of γ rays following Coulomb excitation. The assignments of the 358- and 295-keV levels are $J^\pi = \frac{5}{2}^-$ and $J^\pi = \frac{3}{2}^-$, respectively.

Spin and parity assignments of either $\frac{3}{2}^+$ or $\frac{7}{2}^-$ for the fifth excited state at 537 keV are made in Ref. 1 mainly on the grounds of internal-conversion measurements, while assignments of either $\frac{5}{2}^+$ or $\frac{7}{2}^+$ for this level are made in Ref. 2 on the grounds of γ-ray intensity and directional-correlation studies. Similarly, Ref. 1 assigns the spin and parity of $\frac{7}{2}^-$ to the sixth excited level at 650 keV, while Ref. 2 concludes that the assignments should be either $\frac{7}{2}^+$ or $\frac{5}{2}^+$. The present state of the spin assignments is given in Table I. The internal-conversion and γ-ray intensity studies of the present investigation are generally in agreement with earlier results and lead to parity assignments in agreement with those of Ref. 2, while the much improved accuracy in the 444-53-keV γ-γ directional-correlation measurement of the present investigation allows unambiguous spin assignments for the 93-, 537-, and 650-keV levels (see Fig. 1).

II. EXPERIMENTAL RESULTS

Conversion-Coefficient Measurements

The relative γ-ray intensities were measured using a 33-cm^3 Ge(Li) detector. The detector was calibrated for relative intensity over the energy range from 200 to 700 keV using calibrated sources of ^{110}Ag, ^{133}Ba, and ^{181}Hf. The relative line intensities were also measured in a 4-cm^3 Ge(Li) detector which was independently calibrated for intensity with absolute intensity sources of ^{109}Cd, ^{141}Ce, ^{139}Ce, ^{203}Hg, ^{113}Sn, ^{22}Na, ^{207}Bi, ^{137}Cs, and ^{54}Mn. The two sets of intensities were in good agreement; however, the 444- and 295-keV γ-ray intensity data were far better in the larger-detector experiment.

The internal-conversion electron intensities were measured using a special wide-energy-pass baffle system in a magnetic electron spectrometer of the Gerholm type. For the intensity measurements, the magnetic spectrometer was fitted with a cooled Si(Li) detector at the focus which actually performed the energy discrimination. The current was swept in a linear fashion over the energy region of interest and the pulse-height spectrum of the Si(Li) detector was recorded in a multichannel pulse-height analyzer. The relative efficiency of this system was determined using the sources ^{141}Ce, ^{139}Ce, ^{203}Hg, ^{113}Sn, ^{207}Bi, ^{137}Cs, and ^{54}Mn calibrated for absolute intensity. The transitions of these sources have well-known internal-conversion coefficients, and the absolute e^- intensities were calculated from the γ-ray intensities and the known conversion coefficients. The detection efficiency of this system is found to be very slowly varying with energy, and corrections to the intensities for energy are very small. Generally, the γ-ray and conversion-electron intensities are in agreement with earlier data; hence only the internal-conversion coefficients are presented here. The regions of interest of the e^- spectrum are shown in Fig. 2.

In addition to the relative intensity studies, the internal-conversion coefficient of the K shell and that of the composite line of the $L+M+N+\cdots$ shells of the 497-keV transition, were measured by normalizing to a standard 482-keV γ line in ^{181}Ta. A new measurement of $\alpha_K(482)$ has been averaged with earlier work with the result $\langle\alpha_K(482)\rangle_{av} = 0.0240 \pm 0.007$.[9] This accurately known standard was used to normalize the measurement of $\alpha_K(497)$ in the

TABLE I. Spin-parity assignments for $^{103}_{45}$Rh.

Level energy (keV)	J^π (Ref. 1)	J^π (Ref. 2)
0	$\frac{1}{2}^-$	$\frac{1}{2}^-$
40	$\frac{7}{2}^+$	$\frac{7}{2}^+$
93	$\frac{7}{2}^+$	$\frac{9}{2}^+$
295	$\frac{3}{2}^-$	$\frac{3}{2}^-$
358	$\frac{5}{2}^-$	$\frac{5}{2}^-$
537	$\frac{3}{2}^+$, $\frac{7}{2}^-$	$\frac{5}{2}^+$, $\frac{7}{2}^+$
650	$\frac{7}{2}^-$	$\frac{5}{2}^+$, $\frac{7}{2}^+$

TABLE II. 497-keV internal-conversion coefficient of $^{103}_{45}$Rh.

Authors	α_K
Mei *et al.*[a]	0.0055 ± 0.0003
Shpinel and Kuznetsova[b]	0.006
Zoller *et al.*[c]	0.0043 ± 0.0006
This work	0.0044 ± 0.0004
Theoretical value	
$M1$	0.0046
$E2$	0.0050

[a]J. Y. Mei, C. M. Huddleston, C. M. Allen, and C. G. Mitchell, Phys. Rev. 79, 429 (1950).
[b]V. S. Shpinel and G. A. Kuznetsova, Zh. Eksperim. i Teor. Fiz. 30, 231 (1956) [transl.: Soviet Phys.–JETP 3, 216 (1956)].
[c]See Ref. 2.

present work. The remeasurement of $\alpha_K(497)$ was undertaken in order to reduce the uncertainty in the published values so that this coefficient could be used to normalize conversion coefficients of the 444-, 557-, and 610-keV transitions. The present value of $\alpha_K(497)$ is compared with those of earlier measurements in Table II. In addition the $K/(L+M+N+\cdots)$ ratio was measured in the experiment described above, and was found to be 8.43 ± 0.40 which slightly favors the theoretical value 8.50 for an $M1$ transition over the value of 7.92 for an $E2$ transition. The results of all of these measurements can easily be interpreted, with regard to transition multipolarity, by reference to Fig. 3.

The internal-conversion coefficient of the 53-keV transition was determined by measuring the relative intensities of the 53-keV γ ray and the 20-keV x ray observed in a cooled 200-mm^2-by-2-mm-deep Si(Li) detector. Coincidence with the 557-keV γ ray, observed in a 3×3-in. NaI(Tl) detector, was required. The relative efficiency calibration of the Si(Li) detector was accomplished using a set of sources whose x-ray or γ-ray absolute intensities were measured using a $1\frac{1}{2}\times\frac{1}{4}$-in. NaI(Tl) detector with a 0.005-in. Be window. The lines used for calibration were the K-shell x rays of ^{125}Te, ^{133}Cs, ^{144}Pr, and ^{181}Ta and the 54- and 81-keV γ rays of ^{133}Cs. The results of the internal-conversion-coefficient measurement are compared with earlier results in Table III.

The internal-conversion and γ-ray intensities are generally in agreement with those of earlier work; however, according to our conversion coefficients we find the 610-keV transition to be $M1$, $E2$, or $M1+E2$ but not $E1+M2$ as given in Ref. 1. Furthermore, our conversion-coefficient measurements of the 444-, 497-, and 557-keV transitions imply that these transitions are probably $M1$, $E2$, or $M1+E2$ and not $E1+M2$. We must then assign positive parities to both the 650- and 537-keV levels.

Search for the 242- and the 113-keV γ Rays

The existence of the 242-keV γ ray between the 537 and the 295-keV level with an observable intensity is very important for the assignment of the spin of the 537-keV level. The $M1$, $E2$, or mixed $M1+E2$ character of the 497-keV transition forces us to select $J^\pi(537)=\frac{5}{2}^+$ or $J^\pi(537)=\frac{7}{2}^+$ from the two possible sets of assignments given in Table I. The assignment $J^\pi=\frac{5}{2}^+$ requires the 242-keV γ ray to have a multipolarity of $E1$, while $J^\pi=\frac{7}{2}^+$ requires the multipolarity to be $M2$. Single-particle estimates for the transition probabilities imply the ratio $T(M2)/T(E1)\simeq5\times10^{-8}$; hence an observable intensity of this γ ray would require $J^\pi(537)=\frac{5}{2}^+$.

The search for the 242-keV γ ray was made by

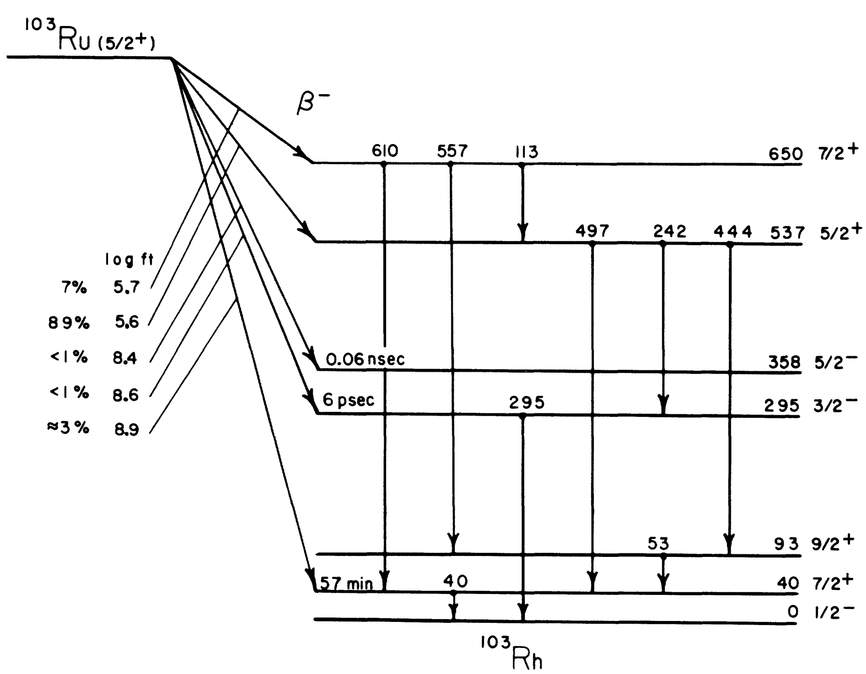

FIG. 1. Decay scheme of ^{103}Rh proposed on the grounds of this and earlier work.

TABLE III. 53-keV internal-conversion coefficient of $^{103}_{45}Rh$.

Authors	α_K
Potnis *et al.*[a]	1.77 ± 0.03
Mukerji *et al.*[b]	2.74 ± 0.03
Sub *et al.*[c]	1.90 ± 0.06
Phelps[d]	2.09 ± 0.04
This work	2.47 ± 0.14
Average value	2.22 ± 0.46
Theoretical value	
$M1$	1.85
$E2$	8.29

[a] V. R. Potnis, E. B. Nieschmidt, C. E. Manderville, L. D. Ellsworth, and G. P. Agin, Phys. Rev. 146, 883 (1966).
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[d] M. E. Phelps, private communication.

TABLE IV. γ-γ directional-correlation measurements ^{103}Rh.

Authors	A_{22} 444-53	A_{22} 557-53
Flack and Mason[a]	0.00 ± 0.02	−0.129 ± 0.020
Singh[b]	0.049 ± 0.013	−0.152 ± 0.011
George *et al.*[c]	0.121 ± 0.060	−0.131 ± 0.014
Zoller *et al.*[d]	−0.16 ± 0.11	−0.17 ± 0.07
This work	−0.23 ± 0.02	−0.130 ± 0.008

[a] F. C. Flack and P. Mason, Proc. Phys. Soc. (London) 71, 247 (1958).
[b] B. P. Singh, Nucl. Phys. 21, 450 (1960).
[c] M. C. George, J. W. Kane, Jr., and A. Mukerji, J. Phys. Soc. Japan, Suppl. 24, 173 (1967).
[d] See Ref. 2.

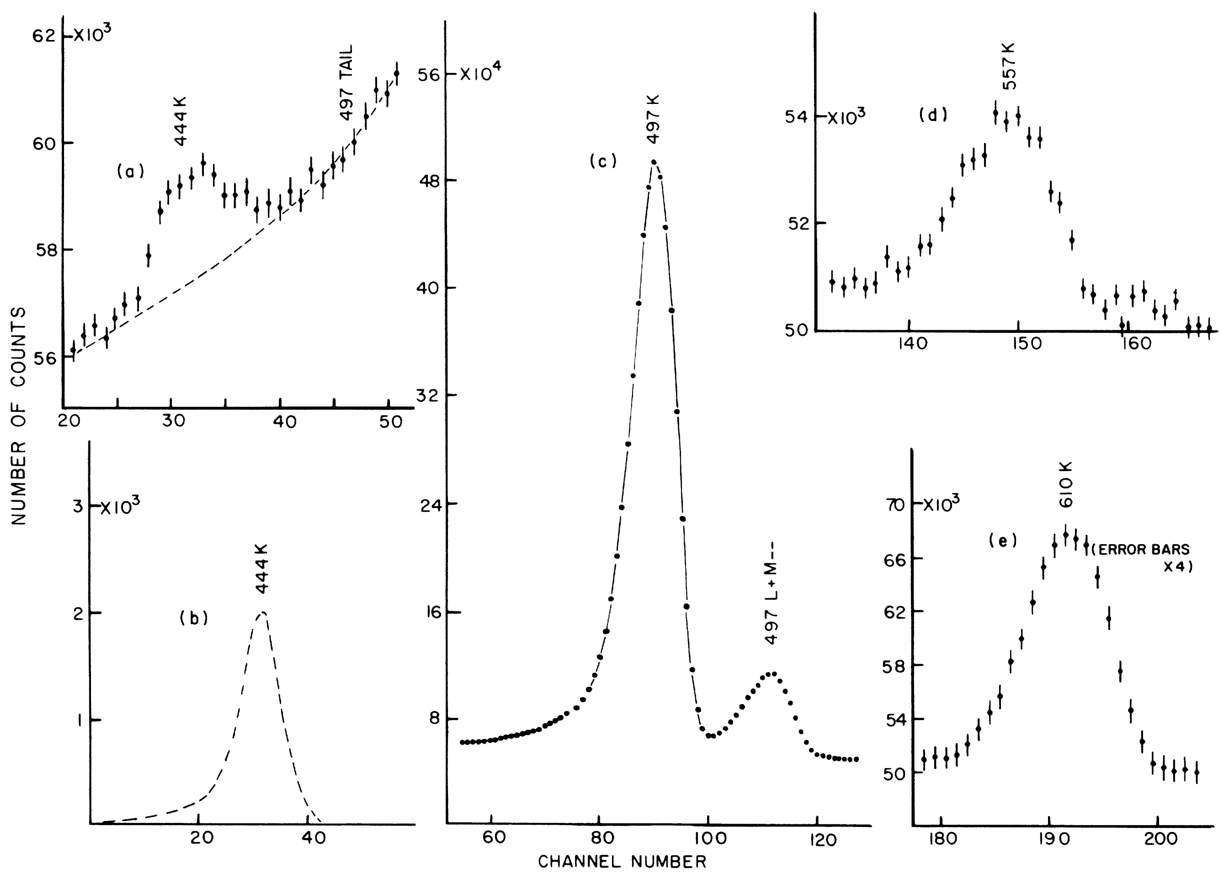

FIG. 2. (a)–(e) Regions of interest of the electron spectrum in the decay of ^{103}Ru. The dashed line in (a) was taken from the tail of the K line of the 482-keV transition in the decay of ^{181}Hf (run under the same conditions). (b) is a mathematical reconstruction of the 444-keV K line after the 497-keV tail is subtracted. The line shape is reasonably good; however, the placement of the dashed line leads to a large error in the area.

placing a weak (~5-μC) source of ^{103}Ru in a close geometry between a 33-cm^3 Ge(Li) detector and a 2×2-in. NaI(Tl) scintillation detector. A timing single-channel analyzer was set to accept a broad region of the γ-ray spectrum of the Ge(Li) channel from about 220 to 275 keV. A second timing single-channel analyzer was set to accept a ~30-keV band centered at 295 keV in the NaI(Tl) spectrum. The energy calibration of the Ge(Li) detector for this experiment was done using the well-known 161-, 223-, 276-, 303-, and 356-keV γ rays from the decay of ^{133}Ba. The calibration of the NaI(Tl) detector was done using the 81-, 303-, and 356-keV γ rays in the same decay and the 511-keV annihilation γ rays from the decay of ^{22}Na. Coincidence pulses were used to gate the multichannel analyzer observing the pulse-height spectrum from the Ge(Li) detector. Several runs of between 48 and 72 h were made both in and out of time alignment and also with the single-channel-analyzer window associated with the NaI(Tl) detector set above and also below the 295-keV γ-ray line. The only experiments which produced a peak at 242 keV in the Ge(Li) spectrum were those with the coincidence apparatus time aligned and with the energy window of the NaI(Tl) channel set near the center of the 295-keV line. The experiment was repeated with the detector axes at 90° and with 3 in. of lead placed between them. The peak at 242 keV was much weaker, because of the increased distance, but was still clearly there. The results of a typical run in the face-to-face detector configuration is shown in Fig. 4. Also shown in Fig. 4 are the results of a similar search for the 113-keV γ ray. In this experiment coincidence was required between the appropriate region of the Ge(Li) pulse-height spectrum and the 497-keV full-energy peak in the NaI(Tl) detector.

We interpret the results of the existence of a 113- as strong verification of the existance of a 113-keV γ ray and a 242-keV γ ray (see Fig. 1). The possibility that these peaks are due to contaminants has been ruled out by searching for similar cascades in all of the possible products which could have been formed in the reactor with isotopically separated ^{102}Ru. It should be mentioned that similar experiments using a ^{103}Ru source from fission products also showed the 242-keV peak; however, the data are of much higher quality with the radioactive source produced via the reaction ^{102}Ru$(n, \gamma)^{103}$Ru.

In light of the earlier discussion, the observable intensity of the 242-keV γ ray implies that this transition is of $E1$ character and requires the assignment $J^{\pi}(537) = \frac{5}{2}^{+}$.

γ-γ Directional-Correlation Measurements

The 557-53 and 444-53-keV γ-γ directional correlations were measured using the 33-cm^3 Ge(Li) detector and a 2×2-in. NaI(Tl) detector mounted on an automatic directional-correlation apparatus. The scintillation detector was used to detect the 53-keV γ ray in both correlation measurements. The 444-53 correlation was measured by frequently cycling the rotating detector and by storing the γ-ray coincidence spectrum in the interval from

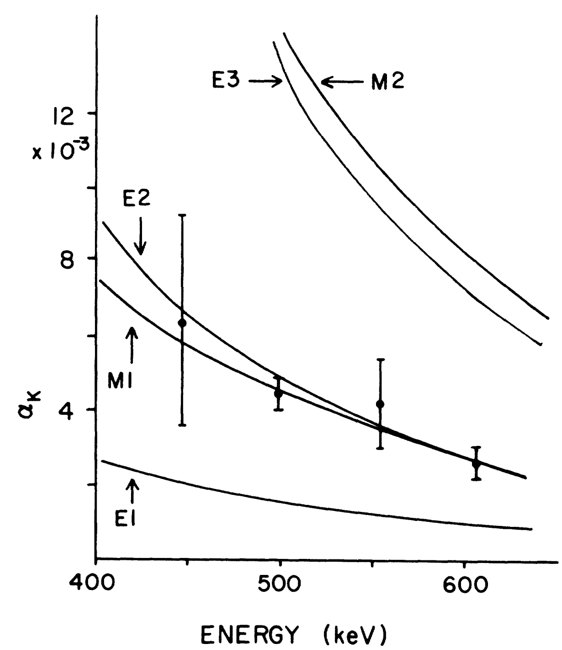

FIG. 3. Comparison of the experimental and theoretical internal-conversion coefficients of the 444-, 497-, 557-, and 610-keV transitions.

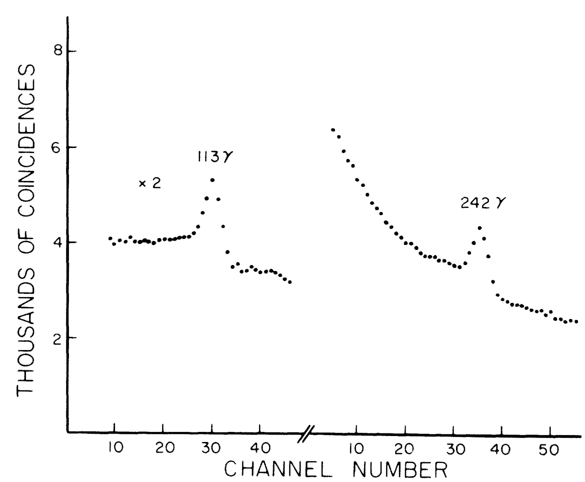

FIG. 4. Weak γ rays in coincidence with the 295-keV γ-ray.

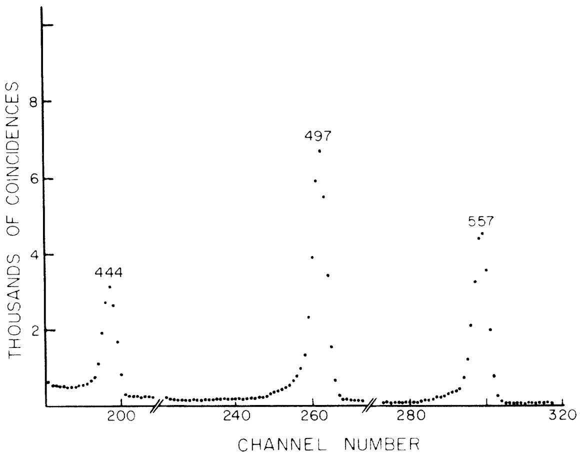

FIG. 5. Typical γ-ray coincidence spectrum from the directional-correlation experiments. The 497-keV peak shown completely vanished after the subtraction of chance coincidences.

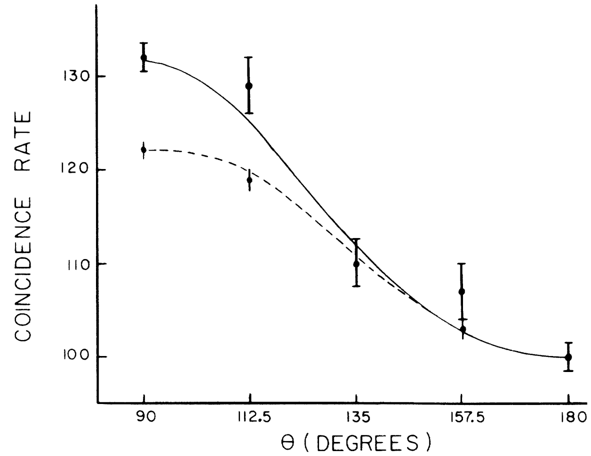

FIG. 6. Experimental correlation functions for the 444-53 (solid line) and the 557-53-keV (dashed line) cascades. The data points for 135 and 180° for the 577-53 cascade lie on those of the 444-53 cascade and were omitted.

400 to 500 keV in a split-memory multichannel analyzer. The γ line was stripped from the continuous background after it was determined that the angular dependence of the background was smooth and had no sharp edges. The 557-53 correlation presents no particular problems and its accurate determination was reasonably straightforward. The 444-53 correlation required almost three months of continuous running in order to achieve the required accuracy. The experimental results of this correlation are shown in Figs. 5 and 6.

The radioactive source used in the correlation measurements was obtained by irradiating isotopically separated ^{102}Ru with thermal neutrons. The 444-53-keV γ-γ correlation data obtained using the present source was quite different from that obtained using a source of ^{103}Ru chemically separated from fission products which invariably contain ^{106}Ru contamination. Interferences from cascades in the decay of ^{106}Ru which result from using the fission-product source were shown to be the cause of our erroneous preliminary results.[10] The results of the directional correlations are compared with those of earlier work in Table IV.

III. DISCUSSION AND CONCLUSIONS

The L-subshell ratios of the 53-keV transition[11] indicate that this transition is very predominantly $M1$ with possibly a small $E2$ mixture. These data indicate that $|\delta| \leq 0.08$ on the grounds of the L_{III}/L_I ratio and $|\delta| \leq 0.12$ on the grounds of the L_{II}/L_I ratio. In addition, the value of the conversion coefficient of the 444-keV transition forces the 444-53 γ-γ cascade to have one of the following spin sequences: $\frac{5}{2} \to \frac{7}{2} \to \frac{7}{2}$, $\frac{7}{2} \to \frac{7}{2} \to \frac{7}{2}$, $\frac{5}{2} \to \frac{9}{2} \to \frac{7}{2}$, or $\frac{7}{2} \to \frac{9}{2} \to \frac{7}{2}$.

The fact that the β branching ratio from the $(\frac{5}{2}^+)$ ground state of ^{103}Ru to the 93-keV level in ^{103}Rh is virtually zero while that to the 40-keV level is well observed, implies that these levels do not have the same spin. The $E1$ nature of the 242-keV γ ray fixes the spin and parity of the 537-keV level to be $\frac{5}{2}^+$. The only spin sequence which explains the present value of $A_{22}(444\text{-}53) = -0.23 \pm 0.02$ and which is in accord with the β branching ratios is the $\frac{5}{2} \to \frac{9}{2} \to \frac{7}{2}$ sequence. This requires that the 444-keV transition be pure $E2$ and implies $\delta(53) = -0.13 \pm 0.03$.

The spin and parity of the 650-keV level is determined above to be either $\frac{5}{2}^+$ or $\frac{7}{2}^+$ on the grounds of the value $\alpha_K(610)$ of the present investigation in agreement with that of Ref. 2. If the spin were $\frac{5}{2}$, then the 557-53 correlation would be identical to the 444-53 correlation. These correlations are certainly not identical, within experimental limits, and the spin of the 650-keV transition is thus determined to be $\frac{7}{2}^+$. The spins of the 93-, 537-, and 650-keV levels are then unambiguously determined to be $\frac{9}{2}^+$, $\frac{5}{2}^+$, and $\frac{7}{2}^+$, respectively. These results imply that the 557-keV transition is a mixed $M1 + E2$ transition with $\delta(557) = -0.32 \pm 0.03$. The phase conventions for the mixing ratios are given in the Appendix.

APPENDIX

Directional-Correlation Conventions

The γ-γ directional-correlation coefficients, $A_{KK} = A_K(1)A_K(2)$, are expressed in terms of the multipole mixing ratios $\delta(1)$ and $\delta(2)$ and F_K coefficients in the following well-known form:

$$A_K(1) = \frac{F_K(L_1 L_1 j_1 j) + 2(-1)^{L_1 + L_1'} \delta(1) F_K(L_1 L_1' j_1 j) + \delta(1)^2 F_K(L_1' L_1' j_1 j)}{1 + \delta(1)^2} ,$$
$$A_K(2) = \frac{F_K(L_2 L_2 j_2 j) + 2\delta(2) F_K(L_2 L_2' j_2 j) + \delta(2)^2 F_K(L_2' L_2' j_2 j)}{1 + \delta(2)^2} , \tag{A1}$$

where L_1 and L_2 are the multipolarities of the first and second radiation, respectively, $L_1' = L_1 + 1$, $L_2' = L_2 + 1$, and j is the spin of the intermediate state in the cascade $j_1(L_1, L_1 + 1) j (L_2, L_2 + 1) j_2$. The numbers 1 and 2 in the expressions $A_K(1, 2)$ and $\delta(1, 2)$ refer to the first and second radiations, respectively.

The multipole mixing ratios of the present work are defined, in terms of emission matrix elements according to the convention proposed by Krane and Steffen,[12]

$$\delta(1) \equiv \langle j \| L_1 + 1 \| j_1 \rangle / \langle j \| L_1 \| j_1 \rangle ,$$

and

$$\delta(2) \equiv \langle j_2 \| L_2 + 1 \| j \rangle / \langle j_2 \| L_2 \| j \rangle . \tag{A2}$$

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