Energy Levels of Rh^{103} from the Decay of Pd^{103} and Ru^{103}†

BABULAL SARAF*
Bartol Research Foundation of The Franklin Institute, Swarthmore, Pennsylvania
(Received October 26, 1954)

The decay schemes of Pd^{103} and Ru^{103} have been studied with the use of two scintillation spectrometers in coincidence. It has been observed that the electron capture activity of Pd^{103} is accompanied by faint γ radiation having the several quantum energies of 40, 65, 300, 365, and 495 kev with absolute intensities per disintegration of 1×10^{-3}, 4×10^{-5}, 1.2×10^{-4}, 6.6×10^{-4}, and 1×10^{-4}. The gamma rays of energy 65 and 300 kev were found to be in coincidence. The inner bremsstrahlung spectrum associated with the capture transition leading to the 57-minute isomeric level in Rh^{103} is of intensity small in comparison with that of the aforementioned gamma rays. The β decay of Ru^{103} is accompanied by gamma rays of energies 55, 440, 495, 555, and 610 kev with absolute intensities per disintegration of 1×10^{-2}, 5×10^{-3}, 0.90, 5×10^{-3}, and 6×10^{-2}. A β spectrum of end point 110 ± 10 kev is coincident with the 610-kev gamma ray. A level scheme for Rh^{103} with probable orbitals has been proposed.

INTRODUCTION

PREVIOUS investigations[1,2] have indicated that Pd^{103} decays by electron capture only to the isomeric state at 40 kev in Rh^{103}. No unconverted quantum radiations were observed. Recently, in an attempt to study the inner bremsstrahlung of the electron capture process, Rietjens, Van Den Bold, and Endt[3] have reported low-intensity gamma quanta (the absolute intensities per disintegration are indicated parenthetically) of energies 503 kev (1.1×10^{-4}), 367 kev (6.0×10^{-4}), 305 kev (1.1×10^{-4}), and 262 kev ($\sim4\times10^{-5}$). By an argument based upon the calculated intensity of the quantum continuum, they estimated the disintegration energy of the electron capture process to be 494_{-12}^{+27} kev. Since Ru^{103} also decays to Rh^{103}, a comparison of the energy levels excited in Rh^{103} with those excited in decay of Pd^{103} should be possible. The gamma rays of Ru^{103} have been extensively investigated previously,[2,4–7] the various sets of results agreeing poorly. Accordingly, the decay schemes of these two radionuclides having a common residual nucleus have been reinvestigated.

PALLADIUM 103

The natural abundance of Pd^{102} is 0.8 percent. An electromagnetically enriched (34 percent) sample of 200 mg of metallic palladium was obtained from Oak Ridge and exposed for 15 days in the Brookhaven reactor. The irradiated material was chemically purified for removal of any rhodium or silver activities. Finally, palladium was precipitated from an alkaline solution by addition of di-methyl glyoxine. In this manner, a pure source of Pd^{103} of strength nearly five millicuries was obtained.

In order to observe any gamma rays or inner bremsstrahlung of low intensity, it was necessary to absorb before the detector the intense 20-kev x-rays of rhodium which accompany K-capture. The gamma-ray spectrum of Pd^{103} as measured with an intervening absorber of surface density 375 mg/cm² of copper between source and detector, is shown in Fig. 1. The full energy peaks of gamma rays of energies 40, 65, 300, 365, and 495 kev are clearly resolved. In the previously mentioned investigation[3] of this activity, the 65-kev radiation appears to have been heavily attenuated in a 1.7-g/cm² thick brass absorber introduced to suppress the intense K x-radiation, and so is undetected.

The bulk of the low-energy pulses of Fig. 1 below 20 volts could not be accounted for as Compton contributions of the high-energy gamma rays. An analysis

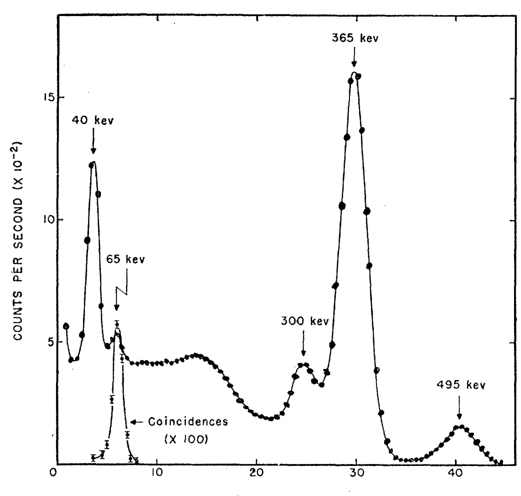

FIG. 1. Gamma-ray spectrum of Pd^{103} as observed in a spectrometer employing NaI(Tl), preabsorption thickness 375 mg/cm² of copper. Coincidences between the 65- and 300-kev gamma rays are also plotted.

† Assisted by the Joint Program of the Office of Naval Research and the U. S. Atomic Energy Commission.

* Research Fellow, Bartol Research Foundation; on leave of absence from Agra College, Agra, India; now at Nobel Institute of Physics, Stockholm, Sweden.

1 D. E. Matthews and M. L. Pool, Phys. Rev. **72**, 163 (1947).
2 Mei, Huddleston, and Mitchell, Phys. Rev. **79**, 429 (1950).
3 Rietjens, Van Den Bold, and Endt, Physica **20**, 107 (1954).
4 C. E. Mandeville and E. Shapiro, Phys. Rev. **77**, 439 (1950).
5 E. Kondaiah, Phys. Rev. **79**, 891 (1950); *M. Siegbahn Commemoration Volume*, p. 442 (Uppsala, 1951).
6 Cork, LeBlanc, Nester, and Stumpf, Phys. Rev. **86**, 575 (1952).
7 F. K. McGowan, Oak Ridge National Laboratory Report ORNL-952, March, 1951 (unpublished).

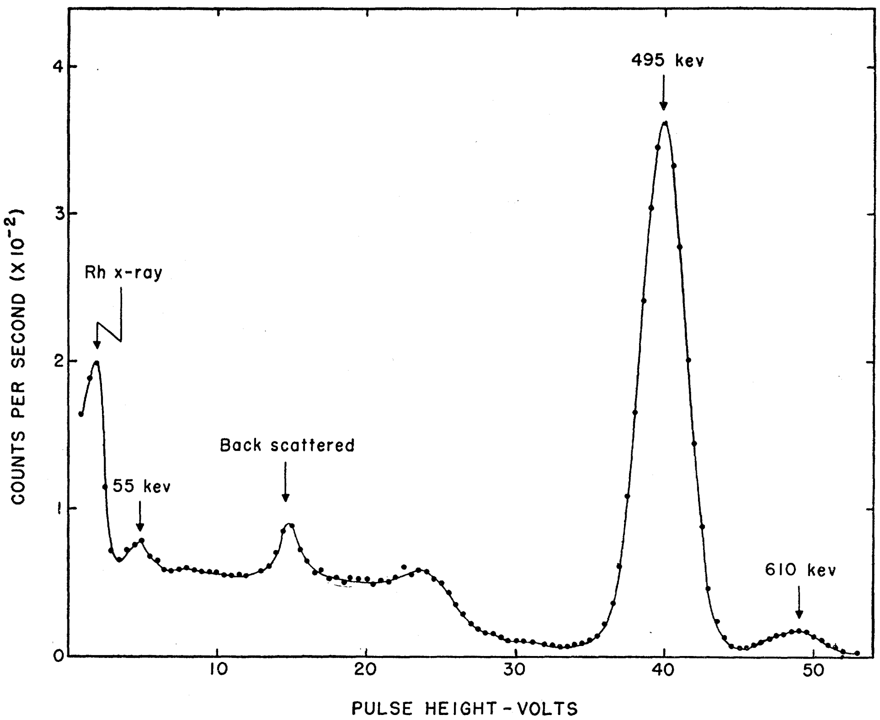

FIG. 2. Gamma-ray spectrum of Ru^{103}.

of the spectrum showed that a gamma-ray continuum of low intensity is partially obscured by the Compton distributions. In addition, the analysis exhibited also the possible presence of a photopeak corresponding to a quantum energy of $\sim$260 kev.[3] This treatment of the data was not so reliable as to yield quantitative information concerning the actual shape of the gamma-ray continuum. However, there was definite indication that the spectral intensity at low energies is considerably greater than that suggested by the early theory of Morrison and Schiff.[8] The spectrum was observed over a period of three months, during which time no significant change in the relative intensities of the various gamma rays was noted.

By a coincidence study, the gamma rays of energies 65 and 300 kev were found to be in cascade. At the lower left of Fig. 1, at a pulse height of 6 volts, a peak corresponding to a gamma-ray energy of 65 kev is present. This photopeak in the coincidence rate was observed by fixing the channel of one spectrometer at 300 kev and scanning the low-energy region with the moving channel of the other spectrometer. No coincidences between any other two gamma rays of the spectrum were detected.

TABLE I. Gamma rays of Pd^{103}.

Probable electron capture transition Energy (kev)	Transition intensity per disintegration	Log ft	Associated gamma-ray energy (kev)	Gamma-ray intensity per disintegration
	$(1.0\pm0.2)\times10^{-4}$		495 ± 5	1×10^{-4}
$\epsilon\sim185$	$(7\pm1)\times10^{-4}$	7.9 ± 0.2	365 ± 5	$(6.6\pm1)\times10^{-4}$
			65 ± 3	$(0.4\pm0.1)\times10^{-4}$
$\epsilon\sim250$	$\sim0.8\times10^{-4}$	9.2 ± 0.2	300 ± 5	$(1.2\pm0.2)\times10^{-4}$
$\epsilon\sim510$	0.999	5.7 ± 0.1	40	$\sim1\times10^{-3}$ (unconverted quanta)

The strength of the source of Pd^{103} was measured by counting the total number of x-rays emitted from the source in a solid angle of 3.75×10^{-3} steradian. The following factors were taken into account: (1) Self absorption in the source and absorption in 40 mg/cm^2 of Al of the crystal assembly; (2) Fluorescence yield of rhodium (0.8); (3) Total conversion coefficient and K/L ratio of the 40-kev radiation. It was further assumed that practically all of the capture transitions terminate at the 57-minute isomeric level in Rh^{103}. The absolute intensities of the gamma rays were estimated from the known thickness of the crystal, the total absorption coefficient for each gamma ray and the ratio for each gamma ray of the area under the full energy peak to the area under the Compton distribution and full energy peak combined. The absolute intensities per disintegration of the various gamma rays are given in Table I along with the associated electron capture transition intensities per disintegration and their respective values of log ft.

The experimental value of the half-life of the transition going to the 535-kev level is approximately 1.5×10^{10} seconds and to the isomeric level 1.47×10^{6} sec. According to Marshak's formula,[9] the mean life of an allowed electron capture transition varies inversely as the square of the decay energy. Thus, the

[8] P. Morrison and L. I. Schiff, Phys. Rev. **58**, 24 (1940).

[9] R. E. Marshak, Phys. Rev. **61**, 431 (1942).

decay energy of an allowed transition leading to the 535-kev level would be about one percent of the energy of the allowed transition to the metastable level. It is probable that the transition to the 535-kev level is indeed allowed. Were it first forbidden, a transition energy of about 100 kev would be necessary to explain the absolute intensity of the 495-kev radiation. Such a transition would in turn lead to an increased energy of the end point of the quantum continuum so that inner bremsstrahlung would be detectable beyond 500 kev, and also the valley between the photopeaks of the 365-kev and 495-kev gamma rays would be less prominent. Hence, in all probability, the electron capture transition to the 535-kev level is allowed.

RUTHENIUM 103

Chemically pure RuO_2 was exposed in the Brookhaven pile to produce the 43-day Ru^{103}. The irradiated material was aged for a month, then further purified for removal of impurities. RuO_4 was distilled from an acid solution in $HClO_4$. The gamma-ray spectrum of Ru^{103} was observed in a cylindrical crystal of NaI(Tl) of diameter 3.5 cm and height 3.5 cm. The data are shown in Fig. 2, where full energy peaks corresponding to quantum energies of 55, 495, and 610 kev are in evidence. A careful comparison in energy was made of the positions of the photopeaks of the 55-kev radiation and the 65-kev gamma rays of Pd^{103}. The difference in energy was found to be 10 ± 2 kev, indicating that two different transitions are involved.

To study beta-gamma coincidences, a thin source of Ru^{103} was placed before an anthracene crystal (dimensions 2.5 cm in diameter, 1 cm thick) which was covered by a reflecting aluminum foil of thickness 1.4 mg/cm^2 and attached to the photomultiplier tube surface with silicone fluid intervening. The gamma rays were detected in NaI(Tl). Curve *A* of Fig. 3 is a plot of the

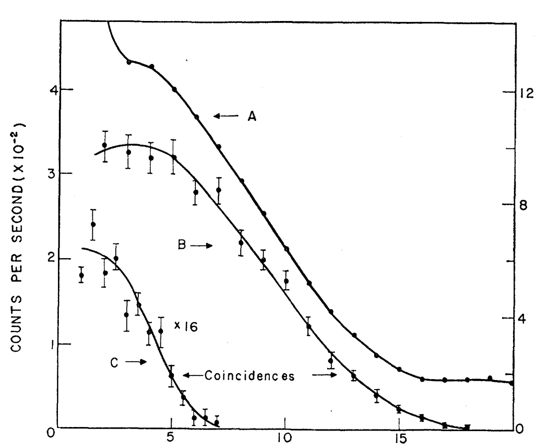

FIG. 3. Curve *A*, pulse-height spectrum in anthracene of an unshielded source of Ru^{103}. Curve *B*, spectrum of beta rays coincident with 495-kev gamma rays. Curve *C*, beta spectrum coincident with 610-kev gamma rays.

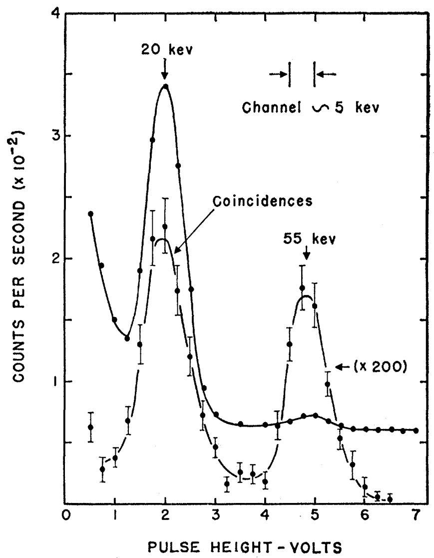

FIG. 4. Spectrum of soft gamma rays coincident with gamma rays of energy greater than 400 kev.

energy distribution of the beta rays of Ru^{103}. Some pulses arising from Compton scattering of the 495- and 610-kev quanta are contained in the spectrum. Beyond a pulse height of twenty volts, the counting rate is undoubtedly composed of both high-energy Compton recoils and hard beta rays from Ru^{103}. Curve *B* of Fig. 3 shows the energy distribution of the beta rays coincident with the 495-kev radiation and curve *C* that of beta rays coincident with the 610-kev gamma rays. Taking the end point of the intense spectrum (Curve *B*) coincident with the 495-kev radiation as 220 kev and taking into account the nonlinear nature of the energy dependence of the pulse heights from anthracene, the end point of the beta spectrum of curve *C* is estimated to be 110 ± 10 kev. This result has also been corrected for energy loss in the covering foil. A search was made for coincidences between the 55-kev radiation and the harder beta rays. From the measurements, it was estimated that at most five percent of the beta rays of the weak hard spectrum might be coincident with any unconverted 55-kev gamma radiation.

In order to determine the nature of the level in de-excitation of which the 55-kev radiation is emitted, gamma-gamma coincidences were studied. In one channel of the coincidence circuit, all pulses corresponding to gamma-ray energies greater than 400 kev were accepted while the channel of the other spectrometer was used to the spectrum at lower energies. A piece of copper, of surface density 1.5 g/cm^2, was placed before the high-energy detector to absorb any *K*-radiation of iodine which might escape to be detected in the low energy scanning channel and thus give rise to spurious coincidences at 30 kev. The coincidence spectrum obtained in this manner is shown in Fig. 4,

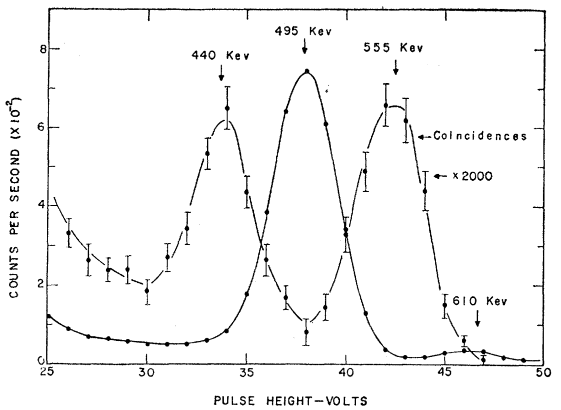

FIG. 5. Spectrum of gamma rays in coincidence with 55-kev radiation.

where a peak of coincidences clearly coincides with a peak at 55 kev in the single counting rate. The peak in the coincidence curve at 20 kev arises from the presence of x-rays of rhodium which result from internal conversion of the 55-kev radiation. A few of the pulses at 20 kev fall in the escape peak of the 55-kev radiation. After correcting for the absorption of the 20 kev and 55-kev radiation in the layer of Al_2O_3 in the crystal assembly and for escape of iodine x-rays[10] in detection of the 55-kev quanta, the K-conversion coefficient of the 55-kev gamma ray was calculated to be 1.2 ± 0.3, indicating a possible $M1$ transition. In similar manner, the coincidence spectrum of Fig. 5 was observed. In this case, pulses corresponding to a gamma-ray energy of 55 kev were accepted in one channel, and the high-energy gamma spectrum was scanned in the other channel. In Fig. 5 are shown coincidence peaks corresponding to gamma-ray energies of 440 and 555 kev. To obtain further assurance that the observed coincidences did indeed correspond to coincidences between the two hard gamma rays and the 55-kev radiation, a molybdenum foil of thickness 550 mg/cm² was placed between the source and the 55-kev detector. The coincidence rates at the peaks of Fig. 5 decreased by more than a factor of 10, as would be expected if the coincidences were actually associated with a 55-kev gamma ray.

Returning to the relative intensities of the gamma rays of Fig. 2, possible branching ratios of the various beta-ray spectra can be estimated, taking the absolute intensity of the 495-kev line as 0.9 per disintegration. The data concerning the β spectra are summarized in Table II.

McGowan[7] has reported gamma rays of energies 312 and 239 kev in Ru^{103}. No unconverted quantum radiations of these energies were detected in the present investigation, suggesting the possibility of presence of impurities in the target material of the earlier studies.[7]

[10] P. Axel, Rev. Sci. Instr. **25**, 391 (1954).

DECAY SCHEMES

The coincidence study of Pd^{103} showed that 65- and 300-kev gamma rays are in cascade. Since the 300-kev quanta are the more intense, it can be concluded that they are preceeded in time by the 65-kev transition. The 365-kev radiation is evidently emitted in the cross-over transition. From all of the previously described measurements, however, it is not possible to ascertain whether the 300 and 365-kev gamma rays terminate at the ground state or the isomeric level at 40 kev. In recent studies of Coulomb excitation of Rh, Heydenburg and Temmer[11] have observed gamma rays of energies 295 and 357 kev. In their measurements, the intensity of the 295-kev radiation is greater than that of the 357-kev line, whereas in the decay of Pd^{103}, the reverse is true. Thus, the two gamma rays cannot originate from the same level, further confirmation of the fact that the 300-kev radiation is preceded by the 65-kev gamma ray. The cross section for Coulomb excitation of the 300-kev gamma ray is such as to suggest that it results from de-excitation of the first rotational level of the ground state. Since the 365-kev radiation is emitted in the cross-over transition of the 65 kev–300 kev cascade, it can be concluded that both the 365-kev and the 300-kev transitions terminate at the ground state of Rh^{103}. A comparison of the energies of the gamma rays from Pd^{103} and from Ru^{103} shows that only the 495-kev gamma ray is emitted in the decay of both radionuclides. The intermediate levels at 300 and 365 kev are not excited in the decay of Ru^{103}.

It has been previously shown[5] that the 495-kev gamma transition of Ru^{103} leads to the 57-minute isomeric level of the residual nucleus so that a level in Rh^{103} is located at 535 kev above the ground state. The energy difference of the end points of the two β spectra, one of which is coincident with the 495-kev radiation and the other with the 610-kev quanta, (see Fig. 3) is 110 ± 10 kev, while the energy difference of the two gamma rays is 115 ± 5 kev, showing that the 610-kev transition must also lead to the isomeric level, originating from a level 650 kev above the ground state of Rh^{103}. From energy considerations, it is con-

TABLE II. Beta spectra and gamma rays of Ru^{103}.

β-ray energy (kev)	Transition intensity per disintegration	Log ft	Associated gamma-ray energy (kev)	Gamma ray intensity per disintegration
110 ± 10	0.07 ± 0.01	5.9 ± 0.2	610 ± 5 555 ± 10	0.06 ± 0.01 0.005 ± 0.002
220	0.90	5.7 ± 0.1	495 ± 5 440 ± 10	0.90 0.005 ± 0.002
670	10^{-3}	10.2	55 ± 3	0.005 ± 0.002
715	0.3	9.3	40	···

[11] N. P. Heydenburg and G. M. Temmer, Phys. Rev. **95**, 861 (1954). The author wishes to thank Dr. Heydenburg for supplying helpful information concerning the spin values of the rotational levels of Rh^{103}.

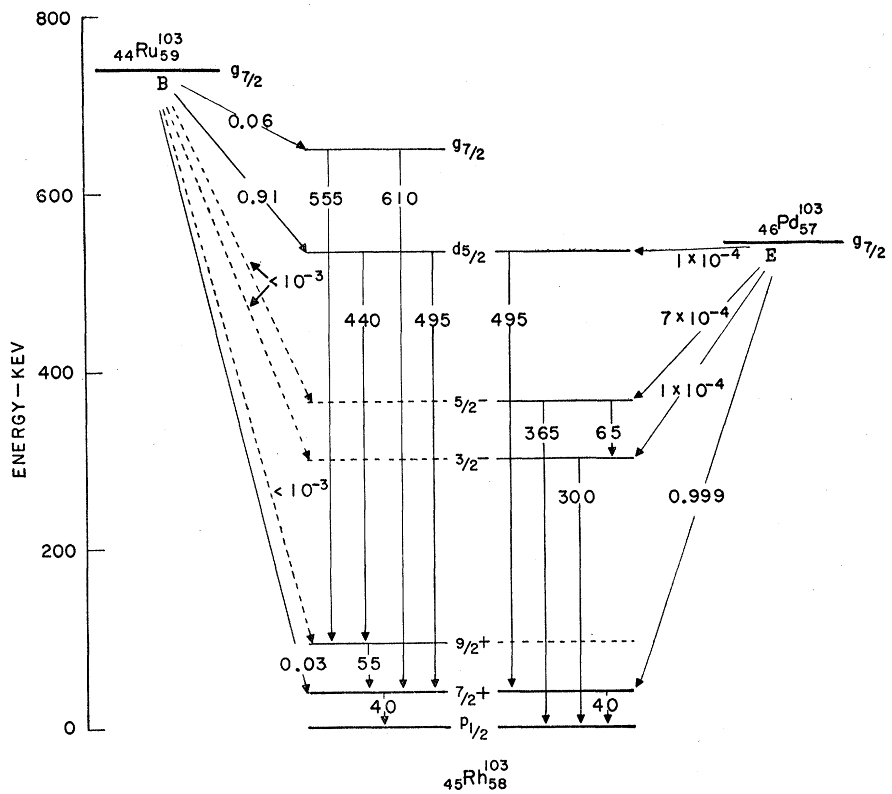

FIG. 6. Energy levels of Rh^{103}

cluded that the aforementioned 495- and 610-kev quanta are emitted in cross-over transitions of the 440 kev −55 kev and 555 kev −55 kev cascades. Thus, a level at 95 kev above the ground state is established.

The spin and magnetic moment of Rh^{103} have been measured.[12] The related orbital was thus determined to be $p_{1/2}$, in agreement with that indicated by the shell model. The 40-kev transition from the 57-minute isomeric level has been classified[13] as $E3$, the level assignment being 7/2+. Since the 55-kev gamma ray is probably emitted in an $M1$ transition; so possible spin values of the level at 95 kev are 5/2+ or 9/2+. The K-shell conversion coefficient of the 495-kev transition is 0.006, the K/L ratio 6.5, and the lifetime less than 2×10^{-9} sec, corresponding to an $M1$ or $E2$ transition[14] ($\Delta I=0$, 1, 2; no!). Thus possible spins for the 535-kev level are 3/2+, 5/2+, 7/2+, 9/2+, or 11/2+. The spin value 3/2+ is not admissible, because there exists no detectable $E1$ transition to the ground state. Since the ground state of Ru^{103} is, according to the shell model, of orbital $d_{5/2}$ or $g_{7/2}$, and since the beta transition between it and the 535 kev level is allowed, the spin of the latter level cannot be 11/2+. A spin of 9/2+ would require that the 7/2+ and 9/2+ configuration states of five $g_{9/2}$ protons have a splitting energy of 495 kev, an improbable condition. The remaining possible spin values for the 535-kev level are 5/2+ or 7/2+. If a purely single particle viewpoint is taken, the level would be characterized by orbitals $d_{5/2}$ or $g_{7/2}$. A similar argument is applicable to the 610-kev gamma transition of the 650-kev level so that its spin assignments could likewise be $d_{5/2}$ or $g_{7/2}$. From a careful consideration of the relative intensities of the 440- and 495-kev gamma rays and the 555- and 610-kev gamma rays, it was concluded that the most probable spin assignments of the 95, 535, and 650 kev levels are 9/2+, $d_{5/2}$, and $g_{7/2}$.

The levels at 300 and 365 kev, which have been interpreted[11] as rotational levels of the collective model of the nucleus, have spin values of 3/2(−) and 5/2(−). If these spin assignments are, indeed, correct, the transition from the 365 kev level to the 57-minute metastable level would be $E1$ and, consequently, prominent, if single particle transition probabilities were to apply. However, no such transition is observed in the present investigations. In the case of Ag, Huus and Lundén[11] have observed a similar $E1$ transition to have only one-fiftieth of the intensity of a competing $E2$ transition. It is further interesting that no observable transitions occur between the single-particle levels at 535 and 610 kev and the supposed rotational levels.

The nucleus of Ru^{103} contains 44 protons and 59 neutrons. According to the shell model, the odd neutron may occupy a $g_{7/2}$ or $d_{5/2}$ state. Either of these orbitals

[12] H. Kuhn and G. K. Woodgate, Proc. Roy. Soc. (London) **A64**, 1090 (1951).
[13] M. Goldhaber and R. D. Hill, Revs. Modern Phys. **24**, 198 (1952).
[14] T. C. Engelder, Phys. Rev. **90**, 259 (1953).

suggests allowed beta transitions to the 535- and 650-kev levels as observed. However, with the invocation of conventional selection rules, it is not possible to explain the large value of log ft of the transition to the 57-minute metastable level ($\Delta I=1$, no, allowed). Recently, De-Shalit and Goldhaber[15] have discussed the problem of anomalous values of log ft for allowed and first forbidden beta transitions. They have emphasized the possible influence of the nuclear core. Their explanation can be applied if the ground state of Ru^{103} is assumed to have orbital $g_{7/2}$. In that event, one or more pairs of neutrons of orbital $d_{5/2}$ will be present in the ground state configuration of Ru^{103}. The most probable configuration would be $g_{7/2}{}^7\ d_{5/2}{}^2$. On the other hand, the effect of the $g_{9/2}$ protons of Rh^{103} on the isomeric and 95-kev levels would be such as to make the neutron configuration of those levels $g_{7/2}{}^8\ d_{5/2}{}^0$. According to De-Shalit and Goldhaber,[15] the beta transitions would be further slowed by the change of orbitals ($d_{5/2}\rightarrow g_{7/2}$) experienced by one pair of neutrons. From these considerations, it can be said that the neutron configuration of the 535- and 650-kev levels of Rh^{103} is probably $g_{7/2}{}^6\ d_{5/2}{}^2$, very similar to the arrangement of the neutron pairs of the ground state of Ru^{103}.

[15] A. De-Shalit and M. Goldhaber, Phys. Rev. **92**, 1211 (1953).

Since the value of log ft for the electron capture decay of Pd^{103} to the isomeric level of Rh^{103} corresponds to an allowed transition, the ground state configuration of Pd^{103} is probably $g_{7/2}{}^7$.

ACKNOWLEDGMENT

The author wishes to acknowledge the kind assistance of C. E. Mandeville both in discussions and in preparation of the manuscript. He wishes also to express appreciation for the continued interest of W. F. G. Swann, Director of the Bartol Research Foundation. The author is also grateful to the Bartol Research Foundation for the award of the fellowship which made possible his stay in the United States.

The author desires also to express his gratitude to N. K. Sethi of Agra College, Agra, India for his kind encouragement. The writer is indebted to the Government of India for award of a travel grant in connection with his visit to the United States.

PHYSICAL REVIEW VOLUME 97, NUMBER 3 FEBRUARY 1, 1955

High-Energy Gamma Rays from the Proton Bombardment of Fluorine*

G. K. Farney,† H. H. Givin,§ B. D. Kern, and T. M. Hahn‡
Department of Physics, University of Kentucky, Lexington, Kentucky

(Received August 19, 1954)

Resonances for emission of gamma rays of energy greater than 8 Mev in the reaction $F^{19}(p,\gamma)Ne^{20}$ have been measured for proton energies from 550 to 1450 kev using a thin target. Resonances were seen at 669, 874, 935, 980, 1090, 1280, 1320, 1355, 1380, and 1430 kev. The effect of false high-energy gamma-ray counts resulting from the nearly simultaneous detection of two 6–7 Mev gamma rays in the reaction $F^{19}(p,\alpha\gamma)O^{16}$ was noted and a method for correcting for this effect was devised. The intensities of gamma rays for the two reactions were compared at each resonance. Also, the angular distribution of the 12-Mev gamma rays emitted at the 669-kev (p,γ) resonance was measured and found to be isotropic to within two percent probable error.

INTRODUCTION

RESONANCES for the emission of 6.1-, 6.9-, and 7.1-Mev gamma rays from the proton bombardment of fluorine in the reaction $F^{19}(p,\alpha\gamma)O^{16}$ are well known.[1] It is also known that at 669-kev proton bombarding energy the compound nucleus Ne^{20*} can decay by alpha-particle emission to O^{16*} and also by gamma-ray emission to the 1.63-Mev level in Ne^{20}. The energy of this gamma ray has been measured to be about 12 Mev.[2,3] It was the purpose of this experiment to look for other resonances for emission of gamma rays of energy greater than 8 Mev, indicating a transition to some lower energy state of Ne^{20}.

The angular distribution with respect to the proton beam of the 12-Mev gamma rays emitted at the 669-kev (p,γ) resonance in fluorine has been measured by using two Geiger counters in an absorption-coincidence measurement.[4] The distribution was found to be isotropic within ten percent. In the present work,

* Sponsored by the Office of Ordnance Research, U. S. Army.
† Now at Bell Telephone Laboratories, Murray Hill, New Jersey.
‡ Now at Virginia Polytechnic Institute, Blacksburg, Virginia.
§ Now at International Business Machines Corporation, Louisville, Kentucky.
[1] F. Ajzenberg and T. Lauritsen, Revs. Modern Phys. **24**, 321 (1952).
[2] Rae, Rutherglen, and Smith, Proc. Phys. Soc. (London) **A63**, 775 (1950).
[3] J. H. Carver and D. H. Wilkinson, Proc. Phys. Soc. (London) **A64**, 199 (1951).
[4] S. Devons and H. G. Hereward, Nature **162**, 331 (1948).