Level scheme of ^{99}Pd and a possible high-spin cascade in ^{99}Rh†

W. F. Piel, Jr., and G. Scharff-Goldhaber
Brookhaven National Laboratory, Upton, New York 11973
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We have verified that a 493.7-, 911.0-, 879.5-, 648.9-, 805.2-, 264.0-keV γ-ray cascade, produced by both $^{12}C+^{91}Zr$ and $^{32}S+^{70}Ge$, occurs in ^{99}Pd by the observation both of coincidences between these γ rays and palladium K x rays and of γ-ray excitation functions. Moreover, this cascade is found to be preceded by a (33.72 ± 0.05)-keV transition. A second γ-ray cascade with side chains also resulting from these bombardments is in addition produced by $^{10}B+^{92}Zr$. Therefore, this second cascade must occur in a nuclide with $Z\leq 45$ and not in ^{99}Pd, as has been proposed by other investigators. Arguments are given in favor of the conjecture that this second γ-ray cascade populates the 4.7-h isomeric state of ^{99}Rh.

[NUCLEAR REACTIONS $^{32}S+^{70}Ge$, $E=110-140$ MeV; measured γ-γ coin, γ-x ray coin, $\sigma_{rel}(E)$, $^{12}C+^{91}Zr$, $E=56-66$ MeV; measured $\sigma_{rel}(E)$, $W(t_\gamma)$ by pulsed beam; $^{10}B+^{92}Zr$, $E=40-55$ MeV; measured $\sigma_{rel}(E)$; ^{99}Pd, ^{99}Rh deduced levels, J^π; enriched targets, Ge(Li), Si(Li) detectors.]

Recently, measurements of γ-ray transitions in ^{99}Pd have been reported.[1] The high-spin states were produced by the reaction $^{90}Zr(^{12}C, 3n\gamma)^{99}Pd$ with $E_{lab}=56$ MeV. The level scheme consisted of two separate γ-ray cascades similar to those obtained by us as shown in Fig. 1. Both cascades were assigned to ^{99}Pd on the basis of γ-ray excitation functions. However, the analysis of the spectrum was made difficult by the simultaneous production of many other end products spanning a range of 4 or 5 for both protons and neutrons.

This communication is concerned with the nuclidic assignment of these two cascades, which we have also observed while studying the neighboring even-even nuclides ^{100}Pd and ^{98}Pd. The cascades were produced at the Brookhaven Tandem Van de Graaff facility by bombarding ^{91}Zr (enriched to 88%) with ^{12}C ions ranging from 56 to 66 MeV and by bombarding ^{70}Ge (enriched to 85%) with ^{32}S ions ranging from 110 to 140 MeV.

During a $^{32}S+^{70}Ge$ experiment at 130 MeV, coincidences between γ rays and x rays were recorded by means of an 80-cm^3 Ge(Li) γ-ray detector located at 90° with respect to the beam axis and a 6.4-cm$^2\times0.3$-cm Si(Li) low energy photon detector, also located at 90° and opposite the γ-ray detector. The Si(Li) detector viewed the ^{70}Ge target through 0.020 cm of beryllium, 0.16 cm of aluminum, and 0.2 cm of Plexiglas. The aluminum was used to attenuate the Ge target K x rays while transmitting the higher-energy K x rays which are of interest, resulting from K shell electron capture or internal conversion in each of the various reaction products.

We observe a previously unreported 33.72-keV transition in coincidence ($2\tau=30$ ns) with all of the other members of the cascade shown to the left in Fig. 1. The third column of Table I lists the relative coincident intensities, corrected for the γ-ray detector efficiency, of the six γ rays in a background-subtracted spectrum gated by γ33.7. The fourth column of Table I lists the relative coincidence rate of Pd K x rays with each of the γ-ray lines. These two columns are normalized to $I=100$ for the coincident Pd K x-ray intensity in the spectrum gated with γ264. In a separate experiment γ-γ coincidences were recorded, resulting from $^{32}S+^{70}Ge$ with $E_{lab}=120$ MeV. The

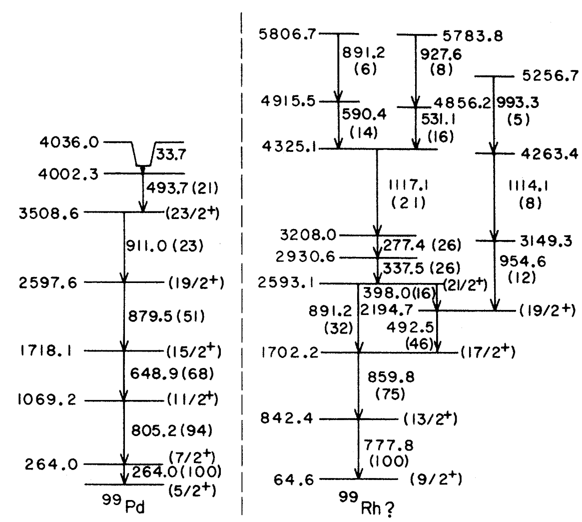

FIG. 1. Two γ-ray cascades previously reported (Ref. 1) to occur in ^{99}Pd. We conjecture that the right-hand cascade with side chains instead takes place in ^{99}Rh. The spins and energies are labeled accordingly. The ordering of the higher-lying transitions in the right-hand cascade is tentative.

TABLE I. Relative intensities of palladium K x rays and of γ33.72 in spectra coincident with background-subtracted gates set on ^{99}Pd transitions produced by the reaction ^{70}Ge(^{32}S,$2pn$)^{99}Pd at 130 MeV. All of the experimental intensities have been corrected for detector efficiencies, electronic triggering efficiencies, and for aluminum absorber transmission. The last two columns list the calculated K x-ray intensities based on the level scheme shown in the left half of Fig. 1, for the two possible multipolarities for γ33.7. These last two columns (best fits taking into account columns 3 and 4) are in moderate agreement with the experimental intensities shown in column 4, for either $E1$ or $M1$ assignment for γ33.7, except for the gate on γ264, as discussed in the text.

Transition energy (keV)	Relative intensity	γ33.72 relative coincident intensity	Pd $K_\alpha + K_\beta$ relative coincident intensity	Calculated relative coincident K x-ray intensity	
				$M1$	$E1$
264.00 ± 0.10	≡100 ± 8	13.6 ± 1.4	≡100 ± 10 [a]	75 [b]	66 [b]
805.2 ± 0.4	94 ± 7	9.9 ± 2.7	90 ± 6	79	86
648.9 ± 0.4	68 ± 14	10.4 ± 2.5	86 ± 5	73	77
879.5 ± 0.4	51 ± 4	13.2 ± 2.9	69 ± 6	69	70
911.0 ± 0.4	23 ± 4	10.3 ± 3.3	57 ± 6	63	60
493.73 ± 0.10	21 ± 2	8.6 ± 1.8	60 ± 4 [c]	74 [d]	70 [d]

[a] Corrected for 25% unresolved Rh K x rays due to an unresolved 264-keV transition (Ref. 4) in ^{99}Rh.
[b] Corrected for the angular distribution (stretched dipole) of γ264.
[c] May contain unresolved (Rh) K x rays due to the other 493-keV transition shown in Fig. 1.
[d] Corrected for the angular correlation of γ493.7.

relative intensities of six γ rays produced by this experiment are shown in the second column of Table I, normalized to $I = 100$ for γ264. The sum of the background-subtracted coincident spectra obtained by gating with each of the six γ rays is shown in Fig. 2.

Figure 3 shows the sum of four background-subtracted Si(Li) spectra gated by γ805, γ879, γ911, and γ494. The 264- and 649-keV transitions are unresolved from other lines, which are unrelated to the cascade under investigation, and are therefore omitted from the sum spectrum. The figure displays the Pd K_α and K_β x rays, as well as the 33.7-keV transition. These data provide the first definitive evidence for assigning the left-hand cascade in Fig. 1 to a Pd nuclide. Constraints on the multipolarity and the relative intensity of γ33.7 are deduced from the data presented in Fig. 3 and Table I as follows. The relative coincidence rates of the 33.7-keV transition and of the Pd K x rays indicate that the 33.7-keV transition precedes the other six γ rays, as shown in Fig. 1. Since the 264-keV transition is known[1] to be $M1$, its K conversion coefficient is $\alpha_K = 0.0242$.[2]

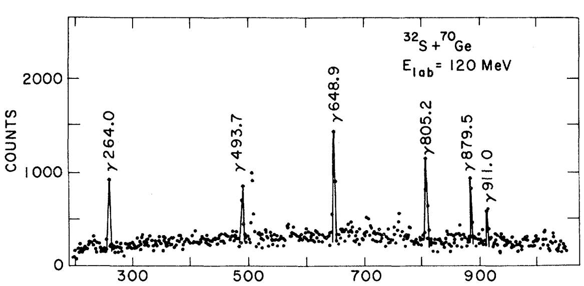

FIG. 2. The sum of the spectra in coincidence with six background-subtracted coincidence gates set on ^{99}Pd transitions, resulting from ^{32}S + ^{70}Ge at 120 MeV. The six γ rays occurring in ^{99}Pd are indicated. Several other peaks, such as γ511, are gated by another unresolved γ264 transition (Ref. 4) in ^{99}Rh.

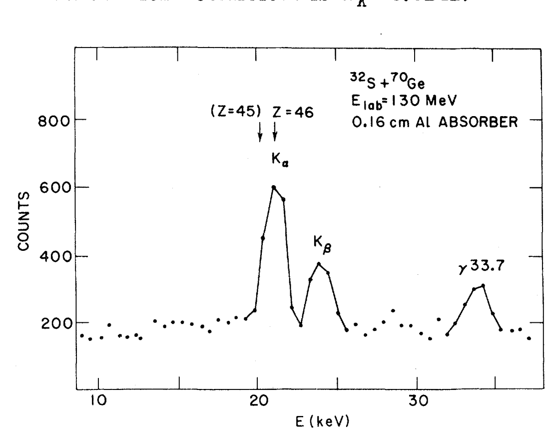

FIG. 3. The sum of the Si(Li) spectra in coincidence with four background-subtracted Ge(Li) coincidence gates set on γ805, γ879, γ911, and γ493 resulting from ^{32}S + ^{70}Ge at 130 MeV. These transitions form the ^{99}Pd cascade shown at the left side of Fig. 1.

From Fig. 3, a ratio of Pd K x rays to 33.7-keV γ rays of 7.1 ± 0.7 is deduced, after correcting for the efficiency of the Si(Li) detector and taking into account the attenuation by the aluminum absorber and the electronic triggering efficiencies. If all of the Pd K x rays in Fig. 3 had resulted from the K shell internal conversion of the 33.7-keV transition, one would deduce a K conversion coefficient for γ33.7 of $\alpha_K^{\text{UNCORR}} = 9.0 \pm 0.9$, using the value $\omega_K = 0.79$ for the Pd K shell fluorescence yield. However, we estimate that at least 5% of the Pd K x rays presented in Fig. 3 are due to the K conversion of γ264. Hence, we arrive at $\alpha_K(\gamma\, 33.7) < 9.4$. This value is to be compared with the K conversion coefficients computed by Hager and Seltzer[2]: 2.77(E1), 7.71(M1), 24.8(E2), and 198(M2). It is seen that the 33.7-keV transition must be essentially a dipole transition. The possibility of an appreciable quadrupole admixture is also ruled out by the lifetime measurement described below. We conclude that $|\delta| < 0.2$, where δ is the quadrupole-dipole mixing ratio. From Fig. 3 and Table I we conclude that the total 33.7-keV transition intensity $N_T = N_\gamma(1 + \alpha_T)$ is ~4% of that of γ264 if γ33.7 is E1, or ~8% if it is M1. The calculated relative coincident Pd K x-ray intensities based on each of these two possibilities are listed in the last two columns of Table I. A comparison of these best fits with the experimental relative coincidence rates presented in column four shows that γ33.7 is more likely M1 than E1. (The fits take into account the data in both columns three and four. The calculated numbers shown in each of the last two columns of Table I are separately scaled to fit column four of Table I in order to facilitate a comparison.) However, since this line was too weak to appear above the background in a Si(Li) singles spectrum, we did not obtain a measurement of its relative intensity.

Lifetime measurements of the observed γ rays were performed by utilizing a pulsed ^{12}C beam and a ^{91}Zr target. An examination of the time-to-amplitude-converter data for γ911 reveals a prompt line shape, from which we conclude that $t_{1/2}(\gamma 33.7) < 4$ ns. The single particle lifetime estimates[3] (corrected for internal conversion[2]) for both E1 (1.3 ps) and M1 (64 ps) satisfy this upper limit, whereas the values for M2 (23 μs) and E2 (2.9 μs) (even an enhanced E2) do not. Hence, as mentioned above, γ33.7 is essentially a dipole transition ($|\delta| < 0.2$).

The decay of 21.4-min ^{99}Pd to levels in ^{99}Rh has been carefully studied by Phelps and Sarantites.[4] The strongest γ rays resulting from this decay are observed in our γ-ray spectra. For the ^{12}C + ^{91}Zr data, we conclude (by assuming equilibrium) that the population of the ^{99}Pd ground state, which results from feeding by the prompt γ-ray cascade ending with γ264, accounts for 58% ($D/1.06A$) of the observed ^{99}Pd ground state decay intensity, as follows from the data presented in Table III and Fig. 4. Similarly, for the ^{32}S + ^{70}Ge data, the feeding is 41%.

Now we consider the second reported γ-ray cascade with side chains terminating with γ777.8, as shown in Fig. 1. We set background-subtracted gates on these transitions, using the ^{32}S + ^{70}Ge data, and looked for coincident K x rays. No definitive K x-ray peak was observed in coincidence with transitions belonging to the second γ-ray cascade nor with its side chains. In order to investigate whether or not the second cascade also occurs in ^{99}Pd, we first compared the relative transition intensities of the two cascades in both the ^{12}C + ^{91}Zr and the ^{32}S + ^{70}Ge bombardments. For this comparison, we set a background-subtracted gate on γ264 and obtained the experimental coincidence intensities if γ805, γ649, γ879, γ911, and γ494 in this spectrum. These intensities are then summed, as shown in Table II, for both bombardments. We should have, in principle, obtained similar results merely from singles intensities. However, the high probability of there being unresolved γ rays in these complex spectra made the simpler method unreliable. Similarly, for the other cascade with side chains we set a background-subtracted gate on γ777.8 and summed the coincident intensities of γ860, γ493, γ398, γ891, γ337, and γ277, after correcting for detector efficiencies. Notice in Table II that the relative population of the two cascades, as measured by the summed coincident γ-ray intensities in each, changes by a factor of 3.44 ± 0.50, although the

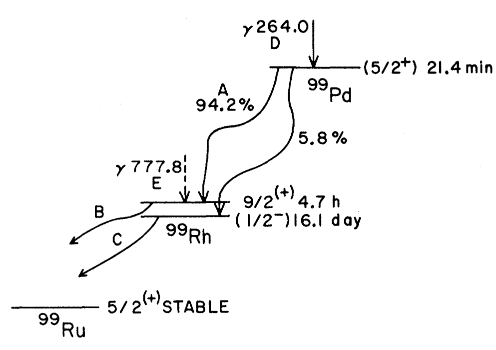

FIG. 4. The part of the $A = 99$ decay chain presently observed. The various total experimental decay intensities are labeled $\boldsymbol{A}$, $\boldsymbol{B}$, and $\boldsymbol{C}$, while the two prompt γ-ray transition intensities are labeled $\boldsymbol{D}$ and $\boldsymbol{E}$. The placement of γ777.8 is based on conjecture. The experimental intensities are listed in Table III and are in agreement with this placement as discussed in the text.

TABLE II. A comparison of relative coincident γ-ray intensities from the bombardments $^{12}C+^{91}Zr$ at 63 MeV and $^{32}S+^{70}Ge$ at 120 MeV. For each γ-ray cascade, the gating transition is the strongest transition. All intensities have been corrected for detector efficiencies. The lower set of transitions are relatively stronger in the $^{12}C+^{91}Zr$ data [(389/101)/(286/256) = 3.44], as discussed in the text.

Transition energy (keV)	$^{12}C+^{91}Zr$	$^{32}S+^{70}Ge$
	Spectrum gated with γ264.0	
805.2	≡94 ± 7	≡94 ± 7
648.9	75 ± 6	68 ± 15
879.5	52 ± 6	50 ± 4
911.0	35 ± 4	23 ± 4
493.7	30 ± 4	21 ± 2
Sum	286 ± 13	256 ± 18
	Spectrum gated with γ777.8	
859.8	157 ± 11	25 ± 5
492.5	81 ± 7	21 ± 5
398.0	25 ± 5	9 ± 6
891.2	55 ± 7	11 ± 4
337.5	35 ± 5	22 ± 3
277.4	36 ± 6	13 ± 3
Sum	389 ± 18	101 ± 11

relative γ-ray intensities within each cascade do not significantly change. This large change of relative intensity in two different bombardments is not characteristic of two parallel high-spin γ-ray cascades occurring in the same nuclide.

To check the possibility that the γ-ray cascade shown to the right in Fig. 1 occurs instead in a nuclide with $Z \leq 45$, we bombarded a $^{92}_{40}Zr$ target (enriched to 95%) with 40, 45, and 55 MeV $^{10}_{5}B$ ions and recorded γ-ray singles spectra. All of the most intense γ rays belonging to the cascade with side chains presented to the right in Fig. 1 were produced. Figure 5 shows the γ-ray singles spectrum obtained at 40 MeV. Moreover, the relative γ-ray intensities are not significantly different from those shown in Fig. 1. Therefore, this second γ-ray cascade *cannot* occur in ^{99}Pd, but must occur in a nuclide with $Z \leq 45$. In Fig. 5 we note that, as expected, there is no evidence for the transitions in the ^{99}Pd cascade shown on the left in Fig. 1. The upper limit for the intensity of the ^{99}Pd cascade is 0.9% of that of the second cascade in the $^{10}B+^{92}Zr$ data.

We conjecture that the cascade at the right in Fig. 1, ending with γ777.8, occurs instead in ^{99}Rh. Although we have no direct evidence for this conclusion, we find that nuclear evaporation calculations based on this hypothesis are in agreement with the observed change of 3.44 ± 0.50 in relative intensities of the two cascades shown in Table II for the $^{12}C+^{91}Zr$ and the $^{32}S+^{70}Ge$ data. Moreover, this conjecture, according to the evaporation calculations, is in agreement with the experimental γ-ray excitation functions for each of the three bombardments: $^{12}C+^{91}Zr$, $E_{lab} = 56-66$ MeV; $^{32}S+^{70}Ge$, $E_{lab} = 110-140$ MeV; and $^{10}B+^{92}Zr$, $E_{lab} = 40-55$ MeV. If the cascade at the right in Fig. 1 would occur instead in an isotope of ruthenium, it would probably already have been found.[5] It is possible that γ777.8 directly populates the 4.7-h isomeric level[4,6] (at 64.6-keV excitation[4]?) in ^{99}Rh which has[7] spin and parity $J^{\pi} = \frac{9}{2}^{(+)}$.

Exploring one additional consequence of this conjecture, we compare in Table III and Fig. 4 the experimental equilibrated intensity E of γ777.8 to the intensities of the known[8] transitions resulting from the decay of the 4.7-h isomer to levels in ^{99}Ru. Notice that if γ777.8 directly populates the 4.7-h isomer in ^{99}Rh, 87% $((A+E)/B)$ of the level population is accounted for in the $^{12}C+^{91}Zr$ data and 68% in the $^{32}S+^{70}Ge$ data. Hence, the conjecture is in agreement with the observed intensity of the 4.7-h ^{99}Rh decay. In both bombardments, equilibrium had been nearly achieved for the decay of the 4.7-h isomer but not for the 16.1-day ^{99}Rh isomer.[8,9] The population of neither level is thought to be near to equilibrium in the $^{10}B+^{92}Zr$ data. Therefore, we have not listed the intensities from this bombardment. All of the spins and parities shown in Fig. 4 are tentative[6] except for the spins of the stable ground state of ^{99}Ru, $J^{\pi} = \frac{5}{2}^{(+)}$, and of the 4.7-h isomer of ^{99}Rh, $J^{\pi} = \frac{9}{2}^{(+)}$, which have been measured.[7,10]

To sum up, we have shown that the γ-ray cascade at the left in Fig. 1 ending with γ264 be-

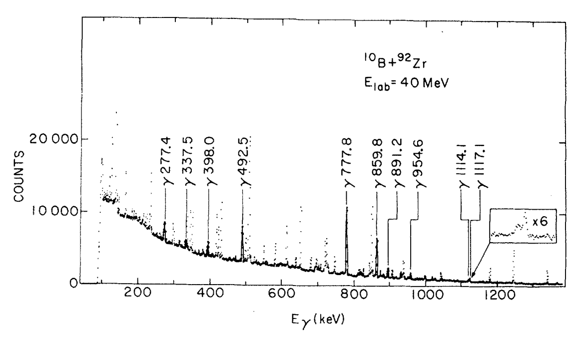

FIG. 5. Ge(Li) γ-ray singles spectrum produced by 40 MeV $^{10}B+^{92}Zr$. The strongest γ rays from the level scheme shown to the right in Fig. 1 are labeled. These transitions cannot occur in ^{99}Pd, but must occur in a nuclide with $Z \leq 45$.

TABLE III. Relative equilibrated intensities for $A=99$ shown in Fig. 4 for $^{12}C+^{91}Zr$ at 63 MeV and $^{32}S+^{70}Ge$ at 120 MeV. The intensities A, B, and C are computed from experimental γ-ray intensities by utilizing known (Refs. 4, 6, 8, and 9) level schemes. The intensity C had not reached equilibrium.

	$^{12}C+^{91}Zr$	$^{32}S+^{70}Ge$
A	162 ± 17	230 ± 25
B	350 ± 30	389 ± 35
C	40 ± 5	<30
D	$\equiv100$	$\equiv100$
E	141 ± 15	35 ± 7

longs to ^{99}Pd on the basis of observed coincident Pd K x rays and of γ-ray excitation functions. The 264.0-keV $M1$ transition is probably a $g_{7/2}\rightarrow d_{5/2}$ single neutron transition, analogous to transitions to the ground states of[1] ^{101}Pd (261 keV) and ^{103}Pd (244 keV), as well as[5] of ^{97}Ru (422 keV), ^{99}Ru (340 keV), and ^{101}Ru (307 keV). We have observed a new low-energy predominantly dipole transition of 33.72 ± 0.05 keV in ^{99}Pd. Moreover, we have shown that a second γ-ray cascade with side chains, previously attributed to ^{99}Pd, does not occur in that nuclide, but occurs in a nuclide with $Z\leq45$. We conjecture that this γ-ray cascade ending with γ777.8 populates the 4.7-h isomer in ^{99}Rh on the basis of γ-ray excitation functions and on the basis of an observed intensity disparity in the $^{12}C+^{91}Zr$ and $^{32}S+^{70}Ge$ data presented in Table II. This conjecture agrees with the observed equilibrated γ-ray intensities which follow[8] β^+ decay or K capture from ^{99}Rh. If this interpretation is correct, it represents the first demonstration of the existence of high-spin states excited in a Rh nuclide.

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