Decay of Sr^{85} and Sr^{85m}

A. W. SUNYAR, J. W. MIHELICH, G. SCHARFF-GOLDHABER, AND M. GOLDHABER,
Brookhaven National Laboratory, Upton, New York,*

AND

N. S. WALL AND M. DEUTSCH, *Laboratory for Nuclear Science and Engineering and Department of Physics, Massachusetts Institute of Technology,† Cambridge, Massachusetts*

(Received February 29, 1952)

Sr^{85} (65 day) decays by K-capture to an excited state of Rb^{85} of 513-kev energy. This state is found to have a half-life of (0.9 ± 0.2) μsec. It decays to the ground state of Rb^{85} by an $M2$ transition, identified by its internal conversion coefficient. Sr^{85m} (70 min) decays in three ways: 14 percent by K-capture to an excited state of Rb^{85} of 150 kev, 84.7 percent by an $E3$ transition of 7.5 ± 1 kev followed by an $M1$ transition of 225 kev, and 1.3 percent by an $M4$ crossover transition of 232.5 kev. A decay scheme for Sr^{85}, Rb^{85m}, and Sr^{85m} consistent with shell theory and isomer systematics is given. The existence of large ft values for allowed β-transitions near closed shells is interpreted as due to a rearrangement of nucleons in the even–even core of an odd A nucleus.

I. INTRODUCTION

IN its ground state Sr^{85} (65 days) decays[1] by orbital electron capture followed by emission of a 513-kev gamma-ray. No positrons have been observed. An isomeric state of 70-minute half-life has been reported to decay with the emission of two gamma-rays[1] of 152 kev and 233 kev. This even–odd isomer which contains 47 neutrons is one of the systematically occurring isomers[2,3] of odd mass number found just before the number of odd nucleons reaches "magic number" 50. In terms of the strong spin-orbit coupling shell model[4,5] $g_{9/2}$ and $p_{1/2}$ levels are here expected to be adjacent in energy. Isomeric transitions in this region should thus be of the $M4$ type ($\Delta I=4$, yes). This prediction is fulfilled in somewhat less than half the cases. The remainder of the isomeric transitions occurring in the $g_{9/2}$ shell have considerably shorter radiation lifetimes (by factors of 10^5 in some cases) and have been identified[6] as $E3$ transitions ($\Delta I=3$, yes), in apparent contradiction to shell-theoretical expectations. This contradiction has been removed[6,7] by a simple extension of the strong spin-orbit coupling shell model, in which it is assumed that an odd number of nucleons (3, 5, or 7) in $g_{9/2}$ states may couple to form a $7/2+$ state which lies lower in energy than the $g_{9/2}$ state. The isomeric transition then takes place between the $7/2+$ state and the $p_{1/2}$ state and is of the $E3$ type. Whenever the ground state of a nucleus is of the $g_{9/2}$ type, it may occasionally happen that the $7/2+$ state lies intermediate in energy between the $p_{1/2}$ and $g_{9/2}$ states. A two-step isomeric transition then becomes possible, $p_{1/2}\rightarrow 7/2+\rightarrow g_{9/2}$, or the transition may proceed competitively via the cross-over $M4$ transition, $p_{1/2}\rightarrow g_{9/2}$. One such example, the isomeric transition in Tc^{99m}, has previously been established.[8]

A number of alternatives had to be considered in the attempts to understand the features of the 70-minute isomeric transition in Sr^{85m}. The isomeric transition may proceed in two steps, or one of the two observed gamma-rays may appear as a result of a competing mode of decay from the isomeric state. One must then decide which gamma-ray is responsible for the lifetime. A comparison of the observed lifetime with the semi-empirical lifetime-energy relation of Goldhaber and Sunyar[6] for $M4$ transitions shows that the 70-minute lifetime is approximately 100 times shorter than expected for an $M4$ transition of 233 kev. If, therefore, we are actually dealing with an $M4$ transition, a competitive process like K-capture must be acting to shorten the lifetime by a large factor. The 152-kev gamma-ray could then be identical with the 149-kev gamma-ray which follows the β-decay from Kr^{85m} and which was identified as a mixed $M1+E2$ transition on the basis of its internal conversion coefficient of 0.051.[9] We also considered the possibility that the isomeric transition is of the $E3$ type, involving a transition between the previously discussed $7/2+$ configuration and a $p_{1/2}$ state, with the 152-kev gamma-ray again appearing in a competitive K-branch. However, when compared to the mean lifetime[6] for all other $E3$ transitions, the Sr^{85m} isomeric transition is slower by a factor of several hundred. When account is taken of the presumed K-branch, the radiative lifetime of the isomeric state is lengthened still further.

A further possibility was that the decay of Sr^{85m} does not involve K-capture, but a two-step isomeric transition between the configurations $p_{1/2}\rightarrow 7/2+\rightarrow g_{9/2}$, with

* Research carried out under contract with the AEC.
† Research supported in part by the joint program of ONR and AEC.

[1] M. Ter-Pogossian and F. T. Porter, Phys. Rev. **81**, 1057 (1951).
[2] E. Feenberg and K. C. Hammack, Phys. Rev. **75**, 1877 (1949).
[3] L. W. Nordheim, Phys. Rev. **75**, 1894 (1949).
[4] M. G. Mayer, Phys. Rev. **78**, 16 (1950).
[5] Haxel, Jensen, and Suess, Z. Physik **128**, 301 (1950).
[6] M. Goldhaber and A. W. Sunyar, Phys. Rev. **83**, 906 (1951).
[7] S. A. Moszkowski, Phys. Rev. **83**, 906 (1951).
[8] Mihelich, Goldhaber, and Wilson, Phys. Rev. **81**, 638 (1951).
[9] I. Bergström and S. Thulin, Phys. Rev. **79**, 537 (1950); I. Bergström, Manne Siegbahn Anniversary volume, Upsala (1951), p. 360.

the 152-kev and the 233-kev gamma-rays emitted in cascade. This situation seemed unlikely for the following reasons: The crossover $M4$ transition would in this case have an energy of 385 kev, which is about the same as that of the isomeric transition in Sr^{87m} (2.7 hr). The remarkable uniformity[6] of the matrix elements of $M4$ transitions makes it appear very likely that the lifetime of such a crossover $M4$ transition in Sr^{85} would be approximately the same as that of Sr^{87m}, since the initial spins would in this case be the same. The 385-kev transition should have been easily observed, unless it was obscured by the presence of Sr^{87m} in the sources used previously. Finally it appeared possible that the 70-minute lifetime is determined by a low energy and as yet unobserved $E3$ transition while the 233-kev gamma-ray is the second step in the cascade. In this case one should also expect the crossover $M4$ transition to occur with an observable intensity. It might have previously escaped detection owing to the fact that the conversion electrons of the 233-kev gamma-ray had not been examined in a spectrometer with sufficient resolution to resolve two close electron groups.

To decide between these possibilities the radiations of Sr^{85m} and Sr^{85} have been studied in detail by various techniques. A unique decay scheme has been established which does involve a complex decay of Sr^{85m} (K-capture, a two-step isomeric transition, as well as a crossover transition). A new short-lived isomer, Rb^{85m}, following K-capture in Sr^{85} has been detected.

II. GROUND-STATE DECAY OF Sr^{85} (65 DAYS)

In order to study the disintegration scheme of the 65-day activity we investigated the coincidences between K x-rays and gamma-rays. The x-rays were detected with an argon-filled proportional counter and single channel pulse-height analyzer. The gamma-rays were detected with a NaI(Tl) scintillation counter and discriminator. By counting coincidences between K x-rays and 513-kev gamma-rays as a function of delay introduced into the x-ray channel, we could establish that the 513-kev state of Rb^{85} is metastable with a half-life of $(9\pm2)\times10^{-7}$ second. By a comparison of the number of 513-kev gamma-rays with the number of x-rays from Sr^{85} we found that substantially all K-capture events lead to the 513-kev state of Rb^{85}.

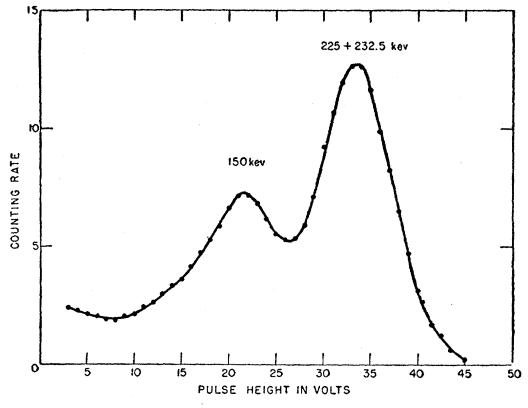

FIG. 1. γ-ray spectrum of Sr^{85m} taken with a scintillation spectrometer [1.4-cm cube NaI(Tl)].

We then measured the internal conversion coefficient of the 513-kev gamma-ray.[10] The result obtained, $\epsilon=(8.1\pm1)\times10^{-3}$, is compatible with either an $M2$ or $E3$ transition,[11] but the measured lifetime excludes an $E3$ transition. The theoretical K conversion coefficient[11] for an $M2$ transition is 6.8×10^{-3}. The empirical K/L ratio curves of Goldhaber and Sunyar[6] indicate that the K/L ratio should be ~9, which leads to a total conversion coefficient of 7.5×10^{-3}. The observed lifetime of 0.9 μsec is slightly higher than the one given by Weisskopf's[12] formula, which yields a value

$$\tau_{M2}(513\text{ kev})=1.6\times10^{-7}\text{ sec.}$$

The conversion coefficient was determined by comparing the number of electrons in the 513-kev line observed in a magnetic lens spectrometer with the number of Rb K x-rays observed in a proportional counter spectrometer. The efficiencies of the two spectrometers for the relevant radiations were calibrated by comparing the corresponding counting rates for Sr^{87m}.

Let N_0 be the number of Sr^{85} atoms decaying per second and $N(e)$ the number of electrons counted in the 500-kev line. $\epsilon=\epsilon_K+\epsilon_L+\cdots$ is the total conversion coefficient and $\eta(e)$ the transmission of the lens spectrometer. Then $N(e)=N_0\epsilon\eta(e)$. Similarly for the x-ray spectrometer, $N(x)=N_0(f_K+\epsilon_K)F\eta(x)$. F is the fluorescence yield for Rb K-shell excitation and f_K is that fraction of electron capture events in which a K electron is captured. Thus $\epsilon=F(f_K+\epsilon_K)N(e)\eta(x)/N(x)\eta(e)$. For Sr^{87m} we denote corresponding quantities with a prime: $N'(e)=N_0'\epsilon'\eta'(e)$ and $N'(x)=N_0'\epsilon_K'F'\eta'(x)$. In this case, of course, observed counting rates must be corrected for decay of the source. Then $N'(x)/N'(e)=(\epsilon_K'/\epsilon')\times F'[\eta'(x)/\eta'(e)]$. Combining this expression with that for ϵ we obtain

$$\epsilon=\frac{N(e)}{N'(e)}\frac{N'(x)}{N(x)}\frac{\epsilon'}{\epsilon_K'}(f_K+\epsilon_K)\frac{F}{F'}\frac{\eta(x)}{\eta'(x)}\frac{\eta'(e)}{\eta(e)}.$$

From the empirical K/L curves[6] we find $\epsilon'/\epsilon_K'=1.15$. We shall not make any serious error if we assume $f_K=0.9$ and $\epsilon_K=0.008$. Thus $(f_K+\epsilon_K)\epsilon'/\epsilon_K'=1.05$. From the standard reference work[13] we find $F/F'=0.97$. The ratio of the x-ray efficiencies involves the prob-

[10] W. S. Emmerich and J. D. Kurbatov [Phys. Rev. **85**, 149 (1952)] have recently come to the same conclusion. In addition, they have found a value of 13 for the K/L ratio of the 513-kev transition. They have measured the internal conversion coefficient of this gamma-ray as 7×10^{-3}. They have been unable to observe K x-ray and gamma-ray coincidences.

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

[12] V. F. Weisskopf, Phys. Rev. **83**, 1073 (1951).

[13] A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand and Company, Inc., New York, 1935).

ability of absorption in the counter gas and the loss in the counter window. From data in the same work[13] we calculate $\eta(x)/\eta'(x)=1.10$. The transmission of the lens spectrometer is substantially independent of energy so that $\eta'(e)/\eta(e)=1$. Thus finally $\epsilon=1.12N(e)N'(x)/N'(e)N(x)$.

Sr^{85} was prepared by deuteron bombardment of rubidium carbonate in the MIT cyclotron. The target was dissolved in dilute acid and the strontium coprecipitated with lead carrier as carbonate. The lead was removed as sulfide from acid solution and the strontium purified further by repeated precipitation as nitrate from fuming nitric acid, using about 0.5 mg of inactive strontium carrier. Y^{87} (80 hr) was prepared by deuteron bombardment of strontium carbonate, and Sr^{87m} was extracted from it periodically by precipitating the nitrate as in the case of Sr^{85}. Sources of both Sr species were prepared by drying a drop containing the radioactive nitrate in an atmosphere of ammonia. This results in a rather uniform deposit of the hydroxide. The ammonium nitrate formed was destroyed by gentle heating. The sources weighed less than 0.3 mg/cm^2 and were deposited on mica weighing about 2 mg/cm^2.

From such sources we obtained $N(e)N'(x)/N'(e)N(x)=7.4\times10^{-3}$, yielding a value for the total conversion coefficient $\epsilon=(8.1\pm1)\times10^{-3}$.

III. DECAY OF Sr^{85m} (70 MIN)

To investigate the radiations from Sr^{85m} this isomer was produced by bombarding Sr^{84} (enriched to 63.7 percent)[14] in the Brookhaven reactor.

We have observed coincidences between K x-rays detected in a proportional counter and gamma-rays detected by a NaI(Tl) scintillation counter. Coincident x-rays characteristic of either Sr or Rb were selected by means of a pulse-height analyzer and critical absorbers. Coincident gamma-ray energies were roughly selected by varying the discrimination level in the scintillation counter. In this manner we could show that the 150-kev gamma-ray but not the 233-kev gamma-ray coincides with K x-rays. The K x-rays coincident with the 150-kev radiation are characteristic of Rb. It is, therefore, clear that the 150-kev radiation appears in a K-electron capture branch from Sr^{85m}, and is very probably the same gamma-ray which is observed to follow β-decay from Kr^{85m}. In the following it will be assumed that these γ-rays are identical.

The intensity ratio of unconverted 233-kev gamma-rays to unconverted 150-kev gamma-rays will yield information about the magnitude of the electron capture branch. This ratio was determined from the pulse-height distribution from a scintillation counter employing a 1.4-cm cube crystal of NaI(Tl). The pulse-height distribution measured with a single channel differential discriminator is shown in Fig. 1. The degree of absorption of the radiations in the crystal was computed from standard reference data.[13] The fraction of the absorbed radiation which appears finally in the photoelectron peak (including multiple Compton effect) was determined by interpolation from experimental data.[15] With these corrections the gamma-ray intensity ratio may be obtained from areas under the photo peaks. This ratio may also be obtained from peak heights of the photoelectron peaks, after additional corrections for variation of the half-width of a photoelectron peak with gamma-ray energy and for change in the relative channel width $\Delta E/E$ with discriminator setting have been made. We obtain the ratio $\gamma_{233}/\gamma_{150}=6\pm2$. This result clearly indicates that the K-branch is relatively small, since the conversion coefficient[9] for the 150-kev gamma-ray is only 0.051. Before the absolute magnitude of the K-branch can be stated, the conversion coefficient for the 233-kev gamma-ray must be determined.

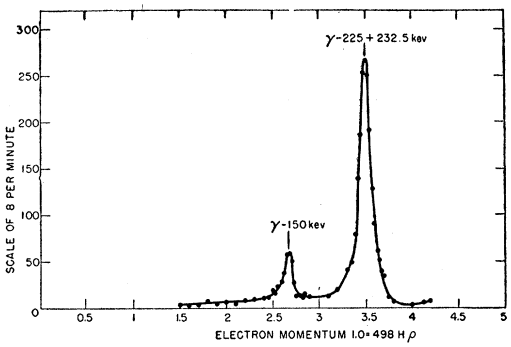

FIG. 2. Internal conversion electrons from Sr^{85m} taken with a lens spectrometer.

This was done by a comparison of the number of conversion electrons from the 150-kev transition with the number of conversion electrons from the 233-kev transition. From the known values of the unconverted gamma-ray intensity ratio and the total conversion coefficient of the 150-kev transition one may then deduce the conversion coefficient for the 233-kev transition. The conversion electron spectrum of the 70-minute activity as measured with a magnetic lens spectrometer is shown in Fig. 2.[16] We obtain a value of 5.0 ± 0.8 for the ratio of the number of conversion electrons of the 233-kev gamma-ray to the number from the 150-kev gamma-ray. If we assume for the moment that the 233-kev transition is simple, this results in a conversion coefficient for the transition of 0.042 which is compatible only with the interpretation that the transition is of an $M1$, $E2$, or mixed $M1+E2$ character. The conversion coefficient is far too small to be compatible with the interpretation that the transition is either an $M4$ or an $E3$ transition. Theoretical K-conversion coefficients[11] for these alternatives are 1.93 and 0.22, respectively, while $\beta_1\sim0.018$ and $\alpha_2\sim0.047$. It is, therefore,

[14] The enriched Sr^{84} was obtained from the Isotopes Division, AEC, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

[15] A. W. Sunyar, unpublished.

[16] We wish to thank Dr. D. E. Alburger for kindly putting his lens spectrometer at our disposal.

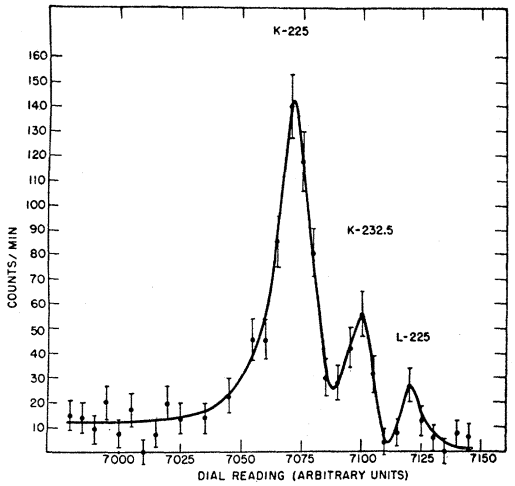

FIG. 3. Internal conversion electrons from Sr^{85m} taken with a 180°-permanent magnet beta-spectrograph and Geiger counter detection. The 225-kev and 232.5-kev transitions are now resolved.

clear that the 233-kev transition cannot be responsible for the 70-minute lifetime.

The approximate value of the conversion coefficient for the 233-kev transition strongly suggests that it is the second step in a $p_{1/2}\rightarrow 7/2+\rightarrow g_{9/2}$ cascade. The energy of the lifetime-determining $p_{1/2}\rightarrow 7/2+$ transition ($E3$) may then be indirectly measured by observation of the crossover $M4$ transition. A search for this transition has been made by examination of the internal conversion electron spectrum of Sr^{85m} in a high resolution, 180° type, fixed magnet spectrograph employing photographic detection. The transition previously called the 233-kev transition was found to be a doublet. K-electron lines of two gamma-rays of 225 and 232.5 kev were observed. The 7.5±1 kev separation of the lines shows that we are not dealing with K and L lines of a single gamma-ray converted in Sr. The 7.5-kev step is accordingly interpreted as the $p_{1/2}\rightarrow 7/2+E3$ transition which is mainly responsible for the 70-minute half-life.

In order to obtain a more reliable intensity ratio of the two K lines than is possible from the weak lines on the film, we have employed detection of electrons with a Geiger counter inside the fixed magnet spectrograph. The Geiger counter with a 1.6-mm entrance slit over the window (2.5 mg/cm² mica) was movable along the film plane by a screw-driven mechanism. The counting rate as a function of position of the Geiger tube along the film plane is shown in Fig. 3. We find that the ratio of the number of K-electrons from the 225-kev gamma-ray to those from the 232.5-kev gamma-ray is 3±1. The K/L ratio for the 225-kev transition is found to be 5.5±2. The K/L ratio expected for the 232.5-kev $M4$ transition from the empirical curve[6] is 5.2. Thus the ratio of the total number of conversion electrons from the 225-kev transition to those from the 232.5-kev transition is also ~3. The number of electrons from the 225-kev transition relative to those from the 150-kev transition then is $5.0\times 3/4=3.75$. The experimentally found ratio $(\gamma_{225}+\gamma_{232.5})/\gamma_{150}=6$ is essentially the ratio $\gamma_{225}/\gamma_{150}$. The total conversion coefficient for the 225-kev transition is $\epsilon_{225}=\epsilon_{150}\times 3.75/6=0.031$. Employing the above K/L ratio we find $\epsilon_K=0.026$ with an estimated uncertainty of a factor of 2. We can, therefore, conclude that the 225-kev transition is either an $M1$, $E2$ or a mixed $M1+E2$ transition. This is in agreement with the interpretation that the transition occurs between a 7/2+ state and a $g_{9/2}$ ground state.

Direct observation of conversion electrons from the lifetime-determining 7.5-kev transition has been attempted by means of absorption measurements in a flow proportional counter. We have observed about 3 times as many soft electrons as can be attributed to the Auger effect. This is below the total number expected by a factor of about 3, a result which can probably be attributed to source absorption effects.

The radiative lifetime for the crossover $M4$ transition may be computed from the available data. Since $\epsilon_K=0.026$ for the 225-kev transition, we have 2.5 K-electrons of this transition appearing for each 100 transitions via the 7.5-kev step, and $2.5/3=0.84$ K-electrons of the 232.5-kev transition. Then the number of crossover transitions $N_c=N_\gamma+N_K+N_L+N_M$. Since $N_\gamma=N_K/\beta_4{}^K=0.84/1.93=0.44$ and $N_L/N_M\sim 3$ for $M4$ transitions, we find $N_c=1.5$ for each 100 transitions of 7.5-kev energy. Similarly, there are 16.5 K-branching events per 100 7.5-kev transitions. This leads to a radiative mean life $\tau_\gamma(232.5\text{ kev})=1.5\times 10^6$ seconds. From the semi-empirical formula[6] for $M4$ transitions for an initial spin of 1/2, the calculated radiative mean life is 1.39×10^6 sec, in good agreement with the observed value.

IV. DISCUSSION

From available experimental data one can now construct a complete disintegration scheme (Fig. 4) involving both ground states and isomeric states of the three isobars Sr^{85}, Rb^{85}, and Kr^{85}. The measured spin and magnetic moment[17] of the ground state of Rb^{85} allow it to be designated as an $f_{5/2}$ state. The spins of Kr^{85m}, Kr^{85} and of the 150-kev state of Rb^{85} have previously been assigned[9] as $p_{1/2}$, $g_{9/2}$ and $p_{3/2}$, respec-

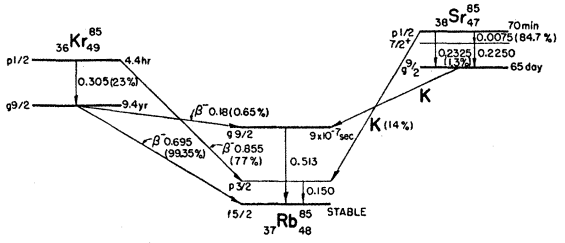

FIG. 4. Proposed disintegration scheme of Kr^{85}, Rb^{85}, and Sr^{85} and their isomers.

[17] National Bureau of Standards Circular 499 (1950).

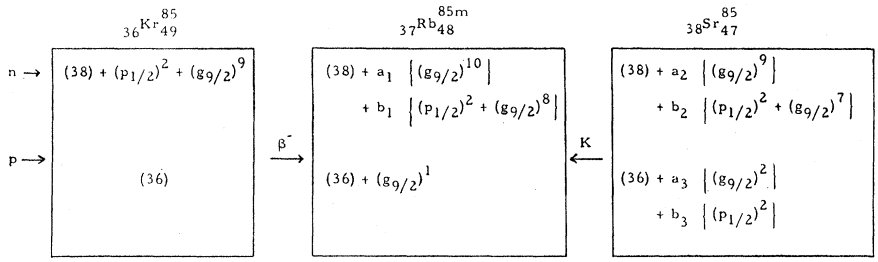

FIG. 5. Suggested neutron and proton configurations for the three isobars Kr^{85}, Rb^{85m}, and Sr^{85}.

tively. We have established that the decay of Sr^{85m} proceeds via a two-step isomeric transition and competitively via the crossover transition between the states $p_{1/2}$, 7/2+ and $g_{9/2}$. The K-capture branch from the isomeric $p_{1/2}$ state of Sr^{85} to the 150-kev state of Rb^{85} is in agreement with the $p_{3/2}$ assignment for the latter state. A search for annihilation radiation was unsuccessful. Positrons, if present, are <15 percent of the K-branch. Using the Feenberg and Trigg curves[18] for the ratio of f_K/f_+ we find a disintegration energy <1.75 Mev for the K-branch. This leads to $ft<10^5$, indicating an allowed transition. For the 65-day Sr^{85} it follows that $ft<5.7\times10^6$, which is also compatible with an allowed transition. The established fact that K-electron capture from the ground state of Sr^{85} does not lead directly to the $f_{5/2}$ ground state of Rb^{85} in any appreciable fraction of the disintegrations, is in agreement with the $g_{9/2}$ designation for the Sr^{85} ground state. Since substantially all such K-capture events lead to the 513-kev excited state of Rb^{85}, and since we have established that the transition to the ground state of Rb^{85} is an $M2$ transition, we must interpret the 513-kev state as a $g_{9/2}$ state. The level assignments from gamma-transitions are thus in excellent agreement with shell-theoretical expectations.

However, an apparent contradiction with beta-theory arises if we consider the beta-emission from Kr^{85}. Zeldes, Ketelle, and Brosi[19] have found that the decay of the ground state of Kr^{85} leads, in 0.65 percent of the disintegrations, to an excited state of Rb^{85}. This state decays by the emission of a 540 ± 20 kev gamma-ray, determined by scintillation spectrometer and absorption techniques. The gamma-ray is in coincidence with the low energy β-group of about 150 kev. We tentatively interpret this gamma-ray as being identical with the 513-kev gamma-ray following K-capture from Sr^{85}, which would, however, imply a slightly higher energy ($\sim$180 kev) for the β-rays which precede it.

Our level assignments have an important bearing upon the relation of comparative half-lives in β-decay to the forbiddenness of the β-transition. The ft value for the 150-kev β-transition from Kr^{85} is of the order of 10^9. The transition takes place between the $g_{9/2}$ ground state of Kr^{85} and the $g_{9/2}$ state of Rb^{85m}. Such a β-transition should be allowed. Accordingly, the ft value would be expected to be of the order of 10^5. Nevertheless, in spite of its allowed character, the transition appears to be slowed down by a factor of the order of 10^4. In the following an explanation for the excessively long half-life of this β-transition is attempted.

The possible neutron and proton configurations for Kr^{85}, Rb^{85m}, and Sr^{85} are shown in Fig. 5. The β-transition from $Kr^{85}\rightarrow Rb^{85m}$ changes a $g_{9/2}$ neutron into a $g_{9/2}$ proton. To the two possibilities for the final neutron configuration of Rb^{85m} are attached probability coefficients "a" and "b". A value of unity for the coefficient b_1 should make the transition proceed as a normal allowed transition. The fact that the transition probability is reduced by a factor of the order of 10^4 is interpreted as evidence that 10 neutrons in $g_{9/2}$ states form a particularly stable configuration preferred to the other possible configuration $(p_{1/2})^2+(g_{9/2})^8$. If the coefficient a_1 were exactly one, the β-transition could not take place, except as a multiple particle transition. The fact that the transition proceeds with $ft\simeq10^9$ indicates that $b\simeq10^{-4}$. The neutron configuration of Rb^{85m} is thus a remarkably pure configuration containing $10g_{9/2}$ neutrons, with an extremely small admixture ($\sim10^{-4}$) of a configuration involving $2p_{1/2}$ neutrons and $8g_{9/2}$ neutrons. Similar shell rearrangements appear to be responsible for large ft values observed in other cases (Zr^{89} and Zr^{89m},[20,21] Y^{87} and Y^{87m}[22]).

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[19] Zeldes, Ketelle, and Brosi, Phys. Rev. **79**, 901 (1950).
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[21] Shore, Bendel, and Becker, Phys. Rev. **83**, 688 (1951).
[22] L. G. Mann and P. Axel, Phys. Rev. **84**, 221 (1951).