Radioactive Decay of Cs^{134} and Cs^{134m}†

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The radiations of Cs^{134} (2.3 yr) and Cs^{134m} (3.15 hr) have been studied in a high-resolution beta-ray spectrometer. The gamma rays of Cs^{134} were observed both by internal and external conversion, and coincidence rates were measured between the beta continuum and the internal conversion electrons of the stronger gamma rays. The beta spectrum of Cs^{134} appears to consist of four, or possibly five, components. Nine gamma rays were found both by internal and external conversion; a tenth gamma ray appears only in internal conversion. A decay scheme is proposed which is reasonably consistent with the multipole order of each of the radiations as obtained from internal conversion data.

A search was made for a ground- or intermediate-state beta transition from Cs^{134m}. No transition to the ground state was observed, but some indication was found for a weak transition to an excited state.

I. INTRODUCTION

THE first detailed study of the radiations from 2.3-year Cs^{134} were made by Elliot and Bell.[1] Since then several additional researches have appeared in the literature,[2] each of which has indicated further complexity in the decay scheme. Waggoner, Moon, and Roberts[3] added three weak high-energy gamma rays to the three strong ones found previously. Schmidt and Keister[4] found that one of the gamma rays was actually two closely-spaced rays, and accordingly proposed a new decay scheme.[5] More recently Cork *et al.*[6] reported a total of eleven gamma rays resulting from the decay of Cs^{134}. These workers proposed still another decay scheme.

We have made a further study of Cs^{134} in an effort to resolve the several inconsistencies in the complicated decay scheme. In this objective we have only been partially successful, for we have yet to find a scheme which is wholly consistent with the experimental measurements. The radiations were examined in a high-resolution ring-focus beta-ray spectrometer.[7] The relative intensities and the conversion coefficients of most of the gamma rays have been determined, the beta spectrum has been resolved into several groups, and coincidences between beta particles and internal conversion electrons have been studied in the spectrometer. These data, together with angular correlation and related measurements,[2] suggest a modified decay scheme for which reasonably consistent spins and parities can be assigned to the various energy levels.

† Supported in part by the U. S. Atomic Energy Commission.
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[1] L. G. Elliot and R. E. Bell, Phys. Rev. **72**, 979 (1947).
[2] K. Way, *Nuclear Data*, National Bureau of Standards Circular No. 499 (U. S. Government Printing Office, Washington, D. C., 1950).
[3] Waggoner, Moon, and Roberts, Phys. Rev. **80**, 420 (1950).
[4] F. H. Schmidt and G. L. Keister, Phys. Rev. **86**, 632 (1952).
[5] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).
[6] Cork, LeBlanc, Nester, Martin, and Price, Phys. Rev. **90**, 444 (1953).
[7] F. H. Schmidt, Rev. Sci. Instr. **23**, 361 (1952). For recent modifications of the instrument, see reference 10.

II. THE GAMMA RAYS AND CONVERSION COEFFICIENTS

The relative intensity of each gamma ray was determined by means of photoelectric conversion in thin thorium foil radiators. The mechanical arrangement of the radioactive material and radiator is shown in Fig. 1. A small Dural capsule containing the active material is made removable, and therefore the same foil and identical geometry can be used with various sources of gamma radiation.

The relative intensity of a photoconversion line in the spectrometer when there is a space distribution of source material and an extended radiator foil will, in general, depend upon the gamma-ray energy in a direct manner, and also in an indirect manner through energy-dependent angle factors.[8] For our particular source and radiator geometry, the dependence of the intensity upon energy through the angle factors is expected to be small in the range 0.5 to 1.5 Mev. In order to verify this conclusion an experimental determination of the

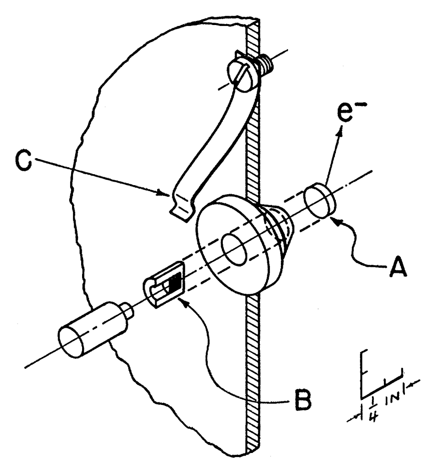

FIG. 1. Photoconversion electron source. (A) Thorium foil radiator; (B) Capsule containing radioactivity; (C) Spring clip to hold source capsule in position.

[8] W. Heitler, *The Quantum Theory of Radiation* (Oxford University Press, London, 1944).

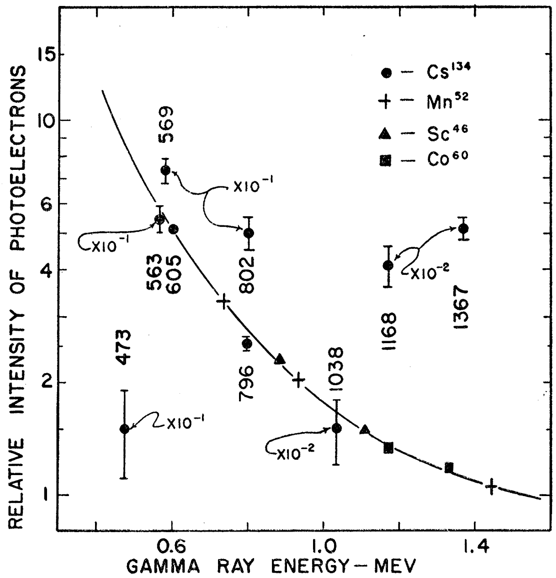

FIG. 2. Gamma-ray intensities of Cs^{134} by photoelectric conversion in thorium foil radiators. The curve is the energy-dependent portion of the theoretical differential cross section.

relative line intensity *vs* energy was made for our particular geometry in the following manner: Sources were studied for which two or more gamma rays are known to be in simple cascade. These include[2] Mn^{52}, Sc^{46}, and Co^{60}, which emit gamma rays of energies that conveniently overlap. The results, as shown in Fig. 2, indicate that the calculated intensity (assuming only the direct energy dependence) is in satisfactory agreement with the measured values over a range from ~0.5 to 1.5 Mev.

High specific activity Cs^{134}, Co^{60}, and Sc^{46} were obtained from Oak Ridge. The Mn^{52} was made in the University of Washington sixty-inch cyclotron.[9]

Of the eleven gamma rays in Cs^{134} reported by Cork *et al.*[6] we were able to resolve and to measure the relative intensities of nine by photoconversion. See Table I. The *K*-shell line of the 663-kev gamma ray,

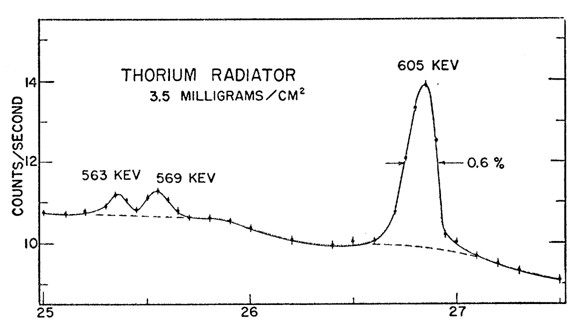

FIG. 3. Typical photoconversion spectrum of Cs^{134} showing the *K*-conversion lines of three of the gamma rays.

[9] Schmidt, Farwell, Henderson, Morgan, and Streib, Rev. Sci. Instr. **25**, 499 (1954).

if present, would be partially obscured by the *L*-shell line due to the 569-kev gamma ray. We were unable to observe the very weak line at 202 kev reported by Cork *et al.*[6] either by external or internal conversion. Typical external conversion spectra are shown in Fig. 3. The relative intensity of each gamma ray, normalized to the 605-kev line, is plotted in Fig. 2.

For study of the beta and internal conversion spectra, thin sources of $CsNO_3$ (specific activity >3 curies/g) were prepared by evaporation from a quartz crucible *in vacuo* onto rubber hydrochloride films of surface density ~0.6 mg/cm². The sources ranged from 16 to 40 micrograms/cm² in average surface density. The uniformity and thinness of the source material is indicated by the absence of any measurable "break" at 600 kev due to Compton electrons produced in the source by the strong 794-kev gamma ray.[10] See Fig. 6.

Several internal conversion spectrum lines are shown in Fig. 4. The spectrometer transmission and luminosity were increased for observation of the weaker gamma

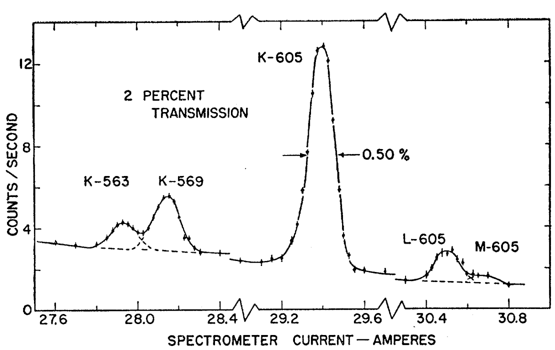

FIG. 4. Representative internal conversion spectrum of Cs^{134}. These lines occur near the end of the continuum. Note that the distance between the lines has been contracted on the graph.

rays. Accordingly, the resolution is not the same for all lines. For the weakest lines the full line width at half-maximum was ~2.4 percent.

The results of the internal and external conversion measurements are summarized in Table I. Our energy values,[11] as tabulated in column 2, are in most cases weighted averages obtained from *K*-shell photoconversion in thorium, *K*-shell internal conversion in barium, and *L*-shell internal conversion in barium. The values published by Cork *et al.*[6] are listed in column 1. Relative intensities of the gamma rays and of the internal conversion lines appear in columns 3 to 6.

Although the evidence is not quite conclusive, both the *K*-*L* energy differences and the photoelectric conversion data for the three high-energy weak gamma rays

[10] G. L. Keister and F. H. Schmidt, Phys. Rev. **93**, 140 (1954). Figure 5 of this paper shows the effect of Compton electrons very clearly.

[11] These differ from the ones published earlier in reference 4. The discrepancy is apparently due to a concurrent new determination of the energy of the *F* line of thorium B which was used as a standard.

support the hypothesis that these gamma rays arise from transitions in barium. This result is in agreement with Cork *et al.*[6] K capture is further excluded by the absence of any measurable Auger electrons. We conclude that a K capture branch cannot exceed one percent. In addition, Mims and Halban[12] placed an upper limit of 0.009 percent on positron emission.

K-shell internal conversion coefficients for each gamma ray are plotted in Fig. 5, and listed in column 2, Table II. These values were obtained by assuming that the 605-kev transition leads to the ground state, and that this radiation is pure $E2$. The coefficient for the 605-kev gamma ray is thus normalized to the theoretical value.[13] We justify the assumption on the following grounds: (1) Angular correlation measurements[14] strongly support a 4-2-0 spin assignment for the levels giving rise to the strongest gamma rays (605 and 796 kev); (2) The coefficient for the 796-kev gamma ray so obtained is thus consistent with an $E2$ assignment; (3) The assignments for other gamma rays is then unambiguous, as can be noted in Fig. 5; (4) The measured K/L ratios are consistent with the extrapolated empirical[15] and theoretical[16] values for $E2$ radiation for both the 605- and the 796-kev gamma rays. The relative gamma-ray intensities and the coincidence measurements (see Sec. IV) support the contention that the 605-kev transition leads to the ground state.

TABLE I. Cs^{134} gamma rays and conversion lines.

Gamma ray energy-kev Cork *et al.*[a]	Present	Relative intensity	Relative intensity of internal conversion electrons K	L	M
202.5	···	<0.2	<1.2		
475.0	473±2	1.8±0.5	3±1		
563.0	563±1	9.4±2.0	11.9±0.6	—[b]	
569.7	569±1	12.8±2.0	26±1	—[b]	
605.4	605±1	100	100[c]	13±1	2.6±0.6
662.7	663±2[d]	···	1.0±0.5		
796.8	796±1	91±4	44±1	6.3±0.6	2.1±0.4
802.6	801±2	18±4	3.9±0.5		
1039	1038±5	0.9±0.2	0.43±0.08	0.06±0.02[e]	
1168	1168±5	3.0±0.4	0.49±0.08	0.08±0.02[e]	
1368	1367±5	4.6±0.3	0.49±0.08	0.05±0.02[e]	

[a] See reference 6.
[b] Cannot be resolved from the K-line of the 605-kev gamma ray.
[c] Corrected for an assumed K/L ratio of ~6 for both the 563- and the 569-kev gamma rays.
[d] Assuming K-conversion in barium.
[e] $L+M$ conversion.

The K-conversion coefficient for the 605-kev gamma ray has also been calculated by assuming that all beta decays lead to this transition. The result, as shown in Fig. 5, gives a coefficient which is ~20 percent less than the theoretical $E2$ value. This is taken as evidence that another gamma ray leads to the ground state in about one out of 5 to 8 disintegrations. The measured intensity of the 801-kev gamma ray indicates that it can play this role.

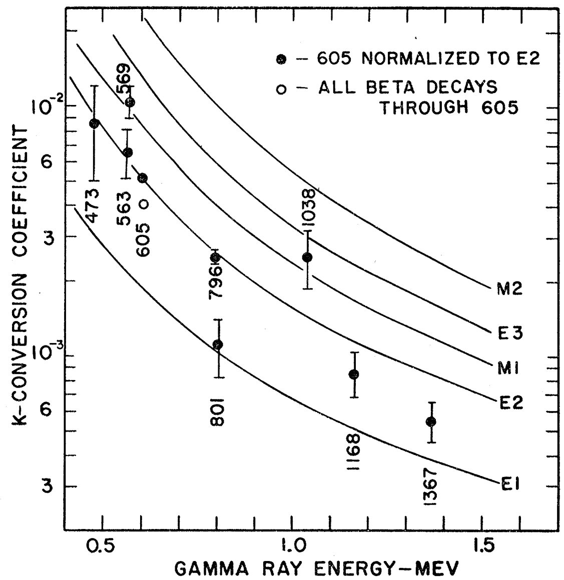

FIG. 5. The K-shell internal conversion coefficients of nine gamma rays in the decay of Cs^{134}. The curves are the theoretical values as obtained from reference 13. The solid points are normalized to the theoretical value for the 605-kev gamma ray. The open circle is the value obtained for the 605-kev gamma if one assumes that all beta decays lead through it.

The other K internal conversion coefficients listed in Table II are plotted in Fig. 5. The solid lines of Fig. 5 are the theoretical[13] values for the various types of radiation. The most probable assignment for each gamma ray, as obtained from Fig. 5, is listed in Table II, column 5.

III. BETA-RAY SPECTRA

The beta-ray continua of both strong and weak Cs^{134} sources (Sec. II) were studied in the spectrometer. The region of the spectrum between the conversion lines which lie very close to the end point was examined at ~1 percent inverse momentum resolution. Detailed measurements of the last 100 kev of the spectrum were

TABLE II. Conversion coefficients of Cs^{134} gamma rays.

Gamma ray energy-kev	K coefficient[a] $\times 10^3$	Experimental K/L	Theoretical[b] K/L_{I+II}	Multipole assignment
473	8.6 ±3.6			$E2$
563	6.6 ±1.5			$E2$
569	10.5 ±1.6			$M1$
605	5.19	7.7±0.6	8.4	$E2$
796	2.51±0.15	8.0±0.8	7.9	$E2$
801	1.1 ±0.3			$E1$
1038	2.5 ±0.7	~ 7[c]		$E3$ or $M1$
1168	0.85±0.18	~ 6[c]		$E1$ or $E2$
1367	0.55±0.10	~10[c]		$E1$ or $E2$

[a] Normalized to theoretical $E2$ value for 605-kev transition.
[b] For $E2$ radiation. See reference 16.
[c] $K/(L+M)$.

[12] W. Mims and H. Halban, Proc. Phys. Soc. (London) **A64**, 311 (1951).
[13] Rose, Goertzel, and Perry, Oak Ridge National Laboratory Report 1023, 1951 (unpublished).
[14] B. L. Robinson and L. Madansky, Phys. Rev. **84**, 604 (1951).
[15] M. Goldhaber and A. W. Sunyar, Phys. Rev. **83**, 906 (1951).
[16] Rose, Goertzel, and Swift, Oak Ridge National Laboratory Report (unpublished). We are indebted to Dr. Rose for kindly supplying us with these data in advance of the full report.

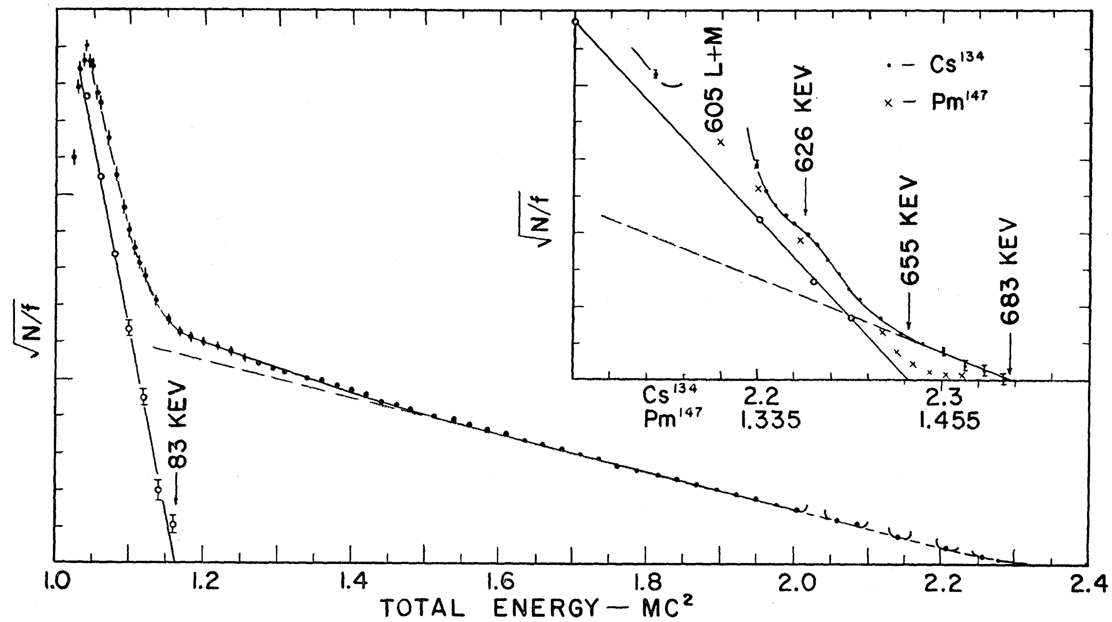

FIG. 6. Fermi-Kurie plot of the Cs^{134} beta spectrum. The region near the end point is shown to expanded scales. A complex spectrum is indicated with end points at 655 and 683 kev. The crosses show the Pm^{147} spectrum shifted to coincide with that of Cs^{134}. The finite resolution of the spectrometer causes a "tailing off" of the Pm^{147} spectrum. A weak internal conversion line appears at 626 kev. The main portion of the spectrum is straight down to about 250 or 300 kev, where one or two weak components may begin. The 83-kev component is clearly evident. It exhibits a straight line Kurie plot down to ~20 kev.

made with a strong source and with the spectrometer baffles set for 4 percent transmission giving an inverse resolution of ~1.7 percent. A Fermi-Kurie plot of the results is shown in Fig. 6. The well-known principal components have end-point energies of 655 and 83 kev. The intermediate region of the spectrum may indicate the existence of two weak components as suggested by Cork *et al.*[6] We have resolved the spectrum into only one additional component in this region.

The inset of Fig. 6 shows the region of the end point of the Cs^{134} spectrum plotted to expanded scales. For comparison purposes the end point of the Pm^{147} spectrum is also shown. At 626 kev there appears a weak conversion line which, if converted in barium, corresponds to a gamma ray of 663 kev. Just to the right of this line there is a definite "break" which may indicate the existence of another component spectrum. There exists the possibility that the *L*-conversion line of the weak 663-kev gamma ray is responsible for the apparent break in the continuum. If this indeed be the case, then

TABLE III. Cs^{134} beta spectra.

End-point energy in kev Cork *et al.*[a]	Present	Percent intensity Cork *et al.*[a]	Present	Log*ft* (present)
	683±4		13±6	9.9
657±5	655±2	81	50	9.2
410±5	308±100	6	5±2	9.0
210		3		
80	83±3	10	32±6	6.4

[a] See reference 6.

the intensity of the *L* line is about equal to the intensity of the *K* line. For a gamma ray of this energy a *K*/*L* ratio of one is very improbable.[15] It is tempting to associate the 663-kev gamma ray with the 662-kev gamma ray of Cs^{137} which has a *K*/*L* conversion ratio[5] of ~4. The effect on the beta continuum due to a Cs^{137} contaminant of amount thus indicated would be negligible. No changes have occurred in the details of the spectrum over a period of four months which could not be accounted for by the 2.3-year half-life of Cs^{134}.

The end-point energies, relative intensities and log*ft* values of the component spectra are tabulated in Table III.

IV. COINCIDENCE MEASUREMENTS

Coincidences between the beta particle continuum and internal conversion electrons were studied in the beta-ray spectrometer with the following apparatus: A trans-stilbene crystal, approximately 0.5 mm thick, was placed about one-half inch behind the beta-ray source. The light from scintillations produced in the crystal were conducted through a 40-inch long Lucite rod to the face of an RCA 5819 photomultiplier tube. The latter was located in a suitable magnetic shield outside the spectrometer vacuum. Only beta particles of energies greater than 90 kev were detected. The lowest energy beta-ray component was therefore not counted.

Another scintillation crystal about $\frac{1}{8}$ inch thick and associated "light piper" was provided at the focus of the spectrometer in place of the customary G-M tube detector. Coincidence pulses between the two photomultipliers were detected by a circuit of resolving time ~0.2 μsec.

The coincidence measurements show that a larger fraction of the 796- than the 605-kev gamma rays follow high-energy beta decays. The measured ratio of 796 to 605 coincidence rates is 1.3±0.2. The measurements show that very few, if any, of either the 563- or the 569-kev gamma rays are in coincidence with high-energy beta particles.

A search was made with NaI(Tl) scintillation counters for delayed gamma-gamma coincidences. None was observed with a half-life greater than 0.2 μsec. However, our measurements do not quite exclude the possibility of a delay in one of the weaker transitions.

V. Cs^{134m}

The spectrum of the 3.15-hour isomer of Cs^{134} was studied in the spectrometer to determine whether beta transitions occur from the isomeric state to one of the excited states in Ba^{134}. Cs^{134m} was made by the $Cs^{133}(d,p)$ reaction in the University of Washington cyclotron.[9] The strength of the beta-ray source was estimated from the known value[17] of the K-conversion coefficient and the measured intensity of the K-conversion line of the 128-kev transition. A very weak continuous spectrum was observed with an end-point energy ~550 kev, and with a relative intensity ~1 percent. No transition to the ground state of Ba^{134} was found.

VI. DISCUSSION OF DECAY SCHEMES

Figures 7 and 8 show, respectively, the decay scheme proposed by Cork *et al.*,[6] and our modified scheme which is somewhat more consistent with our data.

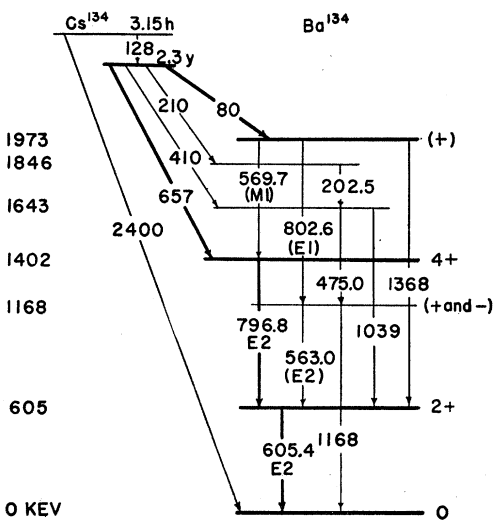

FIG. 7. Decay scheme proposed by Cork *et al.*, reference 6. The multipolarity and parity assignments enclosed in parentheses have been added by us. On the basis of these assignments the level at 1168 kev must be both even and odd.

[17] A. W. Sunyar, Phys. Rev. 83, 864 (1951).

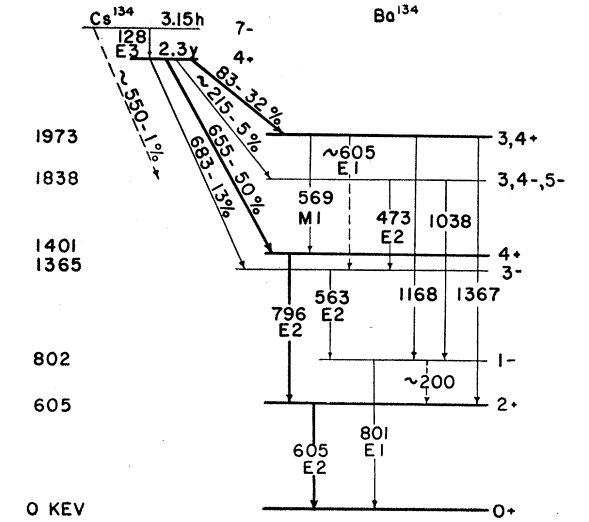

FIG. 8. Newly proposed decay scheme. In order to explain our coincidence and internal conversion measurements, we postulate the existence of a gamma ray of approximately 605-kev energy (shown dotted). The decay scheme for the 3.15-hour isomeric state has recently been shown to consist of a two-step transition [Sunyar, Mihelich, and Goldhaber, Phys. Rev. **95**, 570 (1954)].

The scheme of Fig. 7 is consistent with the observed coincidences between beta particles and internal conversion electrons of the 605- and the 796-kev gamma rays. Also note that, according to this scheme, neither the 563- nor the 569-kev gamma rays are in coincidence with high-energy beta particles. However, about 97 percent of all decays should give rise to the 605-kev gamma ray. The internal conversion coefficient so obtained should then be consistent with $E2$ radiation; as mentioned in Sec. II this is not the case. Further, multipolarity assignments made on the basis of the internal conversion coefficients lead to an inconsistent parity assignment for the second excited state.

The decay scheme shown in Fig. 8 is somewhat more consistent with experimental data than that of Fig. 7. However, it contains a distasteful artifice: In order to understand both the coincidence data and the relative gamma-ray intensities, it seems necessary to postulate the existence of a gamma transition between the 1973- and the 1365-kev levels; its energy would necessarily lie within 2 or 3 kev of the 605-kev gamma, and the conversion coefficient would have to be smaller than that for $E2$ radiation. Further support for this postulate is found by comparing the relative intensities of the 83-kev beta transition with that of the 569-kev gamma ray. The "new" 605-kev gamma ray would thus arise from an $E1$ transition of relative intensity 5 to 15 percent. The existence of the "new" 605-kev gamma would depress the value of the conversion coefficients for the more intense 605-kev gamma ray by less than 10 percent. The still lower value K-conversion coefficient obtained for the 605 when all beta transitions are assumed to pass through the 605 is explained by paralleling the 801 with the 605.

The proposed scheme of Fig. 8 with the newly postulated ~605-kev transition encounters difficulties when one considers the deexcitation of the 1365-kev level by only the 563-kev gamma ray. The situation would be partially alleviated if the 1367-kev gamma ray led from the 1365 level to the ground state. The measured conversion coefficient is then too small to be consistent with the spin assignments. In addition, the coincidence measurements indicate that more low-energy beta decays are followed by 605-kev gammas than by 796-kev gammas. The 1367-kev transition as shown has the appropriate "shunting" effect. For this reason too it is difficult to justify inverting the order of the 796 and 605 gammas, although such an inversion would improve the consistency of the 1038 and 1168 gamma rays.

A ground-state spin of 4 has been measured for Cs^{134} by Bellamy and Smith.[18] An even parity is predicted by the shell model. The principal beta transitions shown in Fig. 8 are therefore allowed and the high logft values must be assumed to result from l-forbiddenness. Similar conclusions apply to the two weaker transitions.

The newly-proposed decay scheme is consistent with the lack of coincidences between high-energy beta particles and the 563-kev internal conversion electrons only if the 683-kev beta component is assumed to be less than ~10 percent of all beta transitions.

We wish to thank T. J. Morgan and the cyclotron crew for the production of Mn^{52} and Cs^{134m}, and J. R. Penning for assistance with the delayed gamma-ray measurements.

[18] E. H. Bellamy and K. F. Smith, Phil. Mag. (7) 44, 33 (1953).

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Deuterium He^3 Reaction*

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By the use of accelerated He^3 ions, the reaction $H^2(He^3,p)He^4$ has been studied in the energy range 100–800 kev (He^3 energy). The angular distribution of the protons was found to be isotropic at bombarding energies of 200, 290, and 350 kev. The reaction cross section has a peak of 695±14 millibarns as determined with reference to the peak value 5.00 barns for the comparison reaction, $H^3(H^2,n)He^4$. The comparison was effected by alternately bombarding the same deuterium target with tritium and He^3 ions, and counting the alpha particles. The peak occurred at 640-kev He^3 ion bombarding energy.

From considerations of the absolute value of the cross section, it is concluded that the resonance at 640 kev is associated with a $J=\frac{3}{2}$ level in the compound nucleus. The experimental shape of the peak is well fitted by resonance parameters in the one-level dispersion formula having the following values: interaction radius R, 5×10^{-13} cm; reduced width for proton emission γ_p, 41.9 kev; reduced scattering width γ_α, 2930 kev; "formal" resonance energy in the center-of-mass system ϵ_s, 391 kev; energy of the level above the ground state of Li^5: 16.2±0.3 Mev. These values agree within experimental error with analogous values associated with the similar resonance which occurs in the $H^2(H^3,n)He^4$ reaction.

INTRODUCTION

The reaction $H^2(He^3,p)He^4$, which we will speak of simply as "the He^3 reaction" is in a sense the mirror image of the $H^2(H^3,n)He^4$ reaction, which we will herein call "the tritium reaction." The former involves Li^5 (three protons and two neutrons) as the intermediate nucleus, and the latter He^5 (two protons and three neutrons). If nuclear forces are charge symmetric, then the two intermediate nuclei should possess energy levels equivalent in character, differing in fact only in Coulomb energy. It is therefore of interest to compare the cross sections of the two reactions as accurately as possible to see if the known resonances have comparable characteristics. It is also of interest to compare the shapes of the resonance peaks with the shapes predicted by the one-level dispersion formula to see if this kind of theoretical treatment can have validity when applied to nuclei possessing as few as five nucleons.

The tritium reaction has been extensively studied in the energy region below 1 Mev,[1–6] with the result that the peak at 163 kev has been characterized in a satisfactory manner both with respect to reduced width and to absolute cross section. The analogous peak in the He^3 reaction is, however, perhaps less well known, although it also has been the subject of a number of

* Submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy at the University of Tennessee.

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