

been assigned a spin and parity of $2-$ or $3+$ by Huus and Day,⁷ and Gove and Paul,⁹ whereas Grant *et al.*⁵ give an assignment of $2+$. No ground state gamma rays have been observed from this level, and an upper limit on the cross section for such transitions has been set at 2.3×10^{-6} barn.⁷ The width of this level is 330 kev.⁷ Using the upper limit on the cross section, an assignment of $2-$ for the spin and parity, and assuming a channel-spin mixture of 0.51 for spins 1 and 2 respectively, the cosine term from interference with this level was calculated to be nine times smaller than that calculated for interference with the 1.4-Mev level.

Interference with the 1.4-Mev level would then seem to provide the better agreement between the calculated and the observed angular distributions. Of the two possible assignments^{7,9} $2+$ and $1-$ previously made for the spin and parity of the 1.4-Mev level, only the latter is consistent with the above results.

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Disintegration of Cs¹³⁸†*

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The decay of the fission product Cs¹³⁸ (32.1 minutes) has been studied with beta- and gamma-scintillation spectrometers and a 141-gauss, 180° permanent-magnet spectrograph. Twelve gamma rays were found with energies of 0.1389, 0.1931, 0.2289, 0.4106, 0.4626, 0.550, 0.87, 1.010, 1.426, 2.21, 2.63, and 3.34 Mev. On the basis of coincidence measurements, intensity data, internal-conversion coefficients, angular correlation data, and the above gamma energies, a consistent decay scheme is proposed which involves levels in Ba¹³⁸ at 1.426, 1.89, 2.21, 2.30, 2.44, 2.63, and 3.34 Mev.

INTRODUCTION

THE radiations emitted by Cs¹³⁸ have been studied by Langer, Duffield, and Stanley¹ and by Thulin.² The former authors reported a total energy release in the decay of 4.8 Mev and found the ground-state to ground-state transition to be of unobservably low intensity. The highest-energy beta group, of end point 3.4 Mev, was shown by beta-gamma coincidence measurements to be followed by a gamma ray of 1.44 Mev. Gamma rays of 0.44 and 0.98 Mev were also found and were shown to lead to the first excited state at 1.44 Mev rather than to the ground state. The beta-ray spectrum was found to be very complex, and its accurate analysis into groups was not possible.

Thulin,² using sources prepared in an electromagnetic separator, examined the gamma radiation with a scintillation counter. He confirmed the existence of the gamma rays reported above, and found others at 0.128, 0.548, 2.24, and 2.68 Mev. No coincidence experiments were done.

In the present work, the investigation of the gamma

radiation following the beta decay of Cs¹³⁸ has been continued with the help of scintillation spectrometers and a permanent-magnet spectrograph. Five more gamma rays have been found, and numerous new coincidence relationships have been established. A consistent decay scheme is proposed.

EXPERIMENTAL METHODS AND RESULTS

Source Preparation

The Cs¹³⁸ used in these experiments came from beta decay of the fission product Xe¹³⁸, which had been made by bombarding U²³⁵ with thermal neutrons from the Los Alamos Water Boiler reactor. The U²³⁵ was irradiated in the form of solid uranyl stearate, UO₂(C₁₈H₃₅O₂)₂, a compound from which the 17-minute Xe¹³⁸ is expected to emanate with ~100% efficiency.³ The uranium compound, contained in a sealed quartz tube, was exposed to a neutron flux of ~6 × 10¹¹ neutrons/cm² sec for 30 minutes, allowed to decay for 10 minutes, and then the fission product gas was transferred to a vessel in which it was allowed to decay for 33 minutes. At the end of this period the remaining gas was pumped out and discarded, and the mixture of cesium and rubidium activities was rinsed off the walls

* Work performed under the auspices of the U. S. Atomic Energy Commission.

† A preliminary report of this work was presented at the Chicago meeting of the American Physical Society [Duffield, Bunker, Mize, and Starnier, Phys. Rev. **100**, 1236(A) (1955)].

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¹ Langer, Duffield, and Stanley, Phys. Rev. **89**, 907(A) (1953).

² S. Thulin, Arkiv Fysik **9**, 137 (1955).

³ Personal communication from Professor A. C. Wahl, of Washington University, who has made a systematic study of the emanating properties of various compounds.

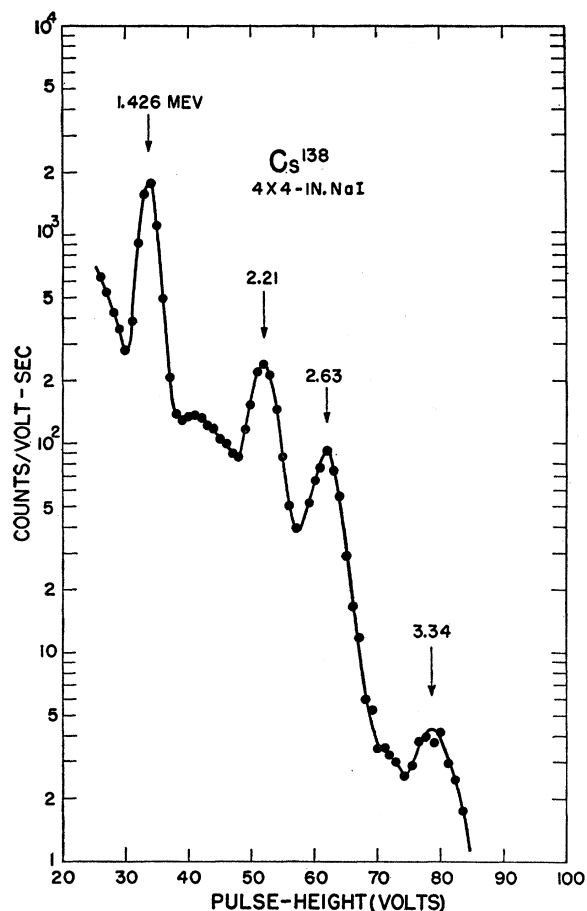


FIG. 1. Pulse-height spectrum of Cs^{138} high-energy gamma rays obtained with a 4×4-inch NaI(Tl) crystal.

of the vessel with water containing cesium and rubidium carrier. The cesium was separated from the rubidium by doing three precipitations of $\text{Cs}_3\text{Bi}_2\text{I}_9$. The activity of the final cesium precipitate was found to decay with a half-life of 32.1 ± 0.2 minutes⁴ over a decay factor range of 10^6 .

The above procedure was used to prepare sources for the gamma-ray singles and gamma-gamma coincidence work. The sources used in the permanent-magnet spectrograph were made by allowing the Xe^{138} to decay in a metal-walled cylindrical vessel of approximately 100 cc volume, along the axis of which passed a stainless steel wire 0.010 inch in diameter. The wire was insulated electrically from the wall and was held at a negative potential of 250 volts with respect to the wall. It was found that approximately one-third of the Cs^{138} ions resulting from the Xe^{138} beta decay were collected on a one-centimeter length of the wire. This portion of the wire was cut off and used directly as a source in the permanent-magnet spectrograph. Since the active de-

⁴ A half-life of 32.2 ± 0.1 minutes has recently been reported by R. M. Bartholomew and A. P. Baerg, *Can. J. Chem.* 34, 201 (1956).

posit was of infinitesimal thickness, good conversion-electron lines were obtained.

The wire sources made in this way contained small amounts of Rb^{88} and Rb^{89} since the fission product gas contained krypton as well as xenon isotopes and no subsequent chemical separation was done. The identity of the conversion-electron lines observed was checked against the gamma rays found in the scintillation work with purified sources containing only Cs^{138} . This point is discussed further below.

Energies and Intensities of Gamma Rays

The gamma radiation was first examined with NaI(Tl) scintillation spectrometers. The crystals were mounted on RCA 6342 photomultiplier tubes, and the amplified output pulses were sorted with a 10-channel pulse-height analyzer. Most of the measurements were made with NaI(Tl) crystals 2 inches in diameter and 2 inches high, but a crystal 1 inch in diameter by $\frac{1}{4}$ inch high and one 4 inches in diameter by 4 inches high were also used for low- and high-energy spectra, respectively. The singles pulse-height spectra which were obtained are shown in Figs. 1 and 2. The energies were determined relative to the gamma energies of the following standards: Na^{22} , Na^{24} , Cs^{137} , Ce^{139} , Co^{60} , Hg^{203} .

The relative intensities of the gamma transitions were calculated from the photopeak areas and an empirical efficiency curve (photopeak area *vs* energy). Since it was known that none of the beta transitions go directly to the ground state of Ba^{138} , the relative gamma-ray intensities could be directly normalized to absolute intensities by referring to the final decay scheme of Fig. 5. These numbers appear in column (2) of Table I. In order to confirm the validity of this

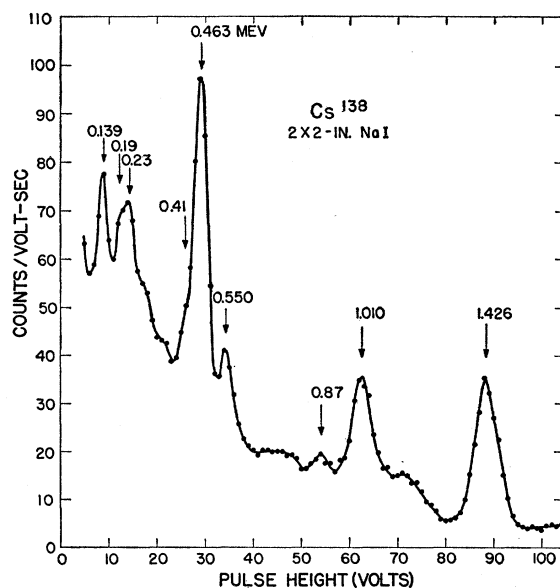


FIG. 2. Pulse-height spectrum of Cs^{138} low-energy gamma rays obtained with a 2×2-inch NaI(Tl) crystal.

normalization, we measured the fraction of the disintegrations which go through the 1.46-Mev gamma ray. (Note: The earlier work¹ had indicated that this intense gamma ray was the transition from the first excited state to the ground state.) The intensity measurement was made as follows: duplicate sources of Cs¹³⁸ and Na²⁴ were prepared, each of total weight ~ 0.2 mg, chemically spread over an area of about 1 cm² on a 50 $\mu\text{g}/\text{cm}^2$ Zapon film. Each source was absolute beta counted in a 4π counter, and the absolute intensities of the Cs¹³⁸ 1.426-Mev gamma ray and of the Na²⁴ 1.368-Mev gamma ray were measured with a calibrated 2 inch by 2 inch NaI(Tl) crystal. From these data, the number of 1.426-Mev gamma rays per beta transition came out to be 0.71 ± 0.05 (value corrected for the fraction of the 1.426-Mev gammas which appeared in a sum peak at 1.89 Mev in the geometry used). This number agrees satisfactorily with the value 0.73 ± 0.07 obtained from the relative gamma-ray intensity measurements.

The gamma intensities reported by Thulin agree approximately with those reported here. Converted to the intensity scale used in Table I, the intensities of the 0.463-, 0.55-, 1.010-, 1.426-, 2.21-, and 2.63-Mev gamma rays found by Thulin were, respectively, 15, 2, 18, 73, 15, and 7.5 percent of the Cs¹³⁸ disintegrations. Thulin also reported a gamma ray at 0.128 Mev which is not

TABLE I. Cs¹³⁸: gamma-ray energies and intensities from scintillation data.

(1) Gamma energy ^a (Mev)	(2) Gamma intensity (percent of Cs ¹³⁸ disintegrations)	(3) Gamma rays verified to be coincident (or non- coincident) with tran- sition in column (1) (Mev)
0.139 \pm 0.003	2 \pm 0.6	0.463, 0.87, 1.426
0.19 \pm 0.01	0.8 \pm 0.3	...
0.23 \pm 0.01	1.6 \pm 0.5	2.21, (not 2.63)
0.410 \pm 0.005	3 \pm 1	0.463, 1.426
0.463 \pm 0.003	23 \pm 3	0.139, 0.410, 0.550, 1.426, (not 1.010)
0.550 \pm 0.006	8 \pm 1.5	0.463, 1.426
0.87 \pm 0.01	4 \pm 1	0.139, 1.426
1.010 \pm 0.006	25 \pm 3	1.426, (not 0.463)
1.426 \pm 0.005	73 \pm 7	0.139, 0.410, 0.463, 0.550, 0.87, 1.010, (not 2.21 or 2.63)
2.21 \pm 0.015	18 \pm 2	0.23, (not 1.426)
2.63 \pm 0.015	9 \pm 1	(not 0.23 or 1.426)
3.34 \pm 0.04	0.5 \pm 0.2	...

^a More precise energy values for some of these gammas are listed in Table II.

to be identified with the 0.139-Mev gamma in Table I because it is approximately ten times too intense. A possible explanation is that the 0.128-Mev line accompanies the beta decay of the 17-minute Xe¹³⁸ parent of Cs¹³⁸. The sources used by Thulin contained a mixture of the two activities, and the gamma lines were assigned to Xe or Cs by following their decay.

Gamma-Gamma and Beta-Gamma Coincidence Measurements

Gamma-gamma coincidence experiments were carried out with a coincidence spectrometer previously described.⁵ The resolving time was $2\tau = 0.4$ μsec . The spectrum coincident with pulses in a particular photopeak of interest was displayed on a slow 100-channel analyzer (dead-time: ~ 67 milliseconds). Figures 3 and 4 show representative data. Figure 3 shows a portion of the singles spectrum and also the spectrum in coincidence with the 0.43- to 0.52-Mev region. Figure 4 shows a different portion of the spectrum, both ungated and gated with pulses in the 2.21-Mev photopeak. The possibility of misinterpretation of coincidence data due to contamination of the selected gate-energy region by Compton pulses of higher-energy gamma rays was appreciated and avoided by observing the effect on the coincidence spectrum of moving the gate interval to a position slightly above the selected photopeak. Where necessary, precautions both as to geometrical arrangement and shielding were taken in order to eliminate possible coincidences resulting from Compton-scattered quanta. A summary of the verified coincidences appears in column (3) of Table I.

A few observations were made of the beta spectrum in coincidence with selected gamma-ray photopeaks. A plastic scintillator was used for detecting the beta

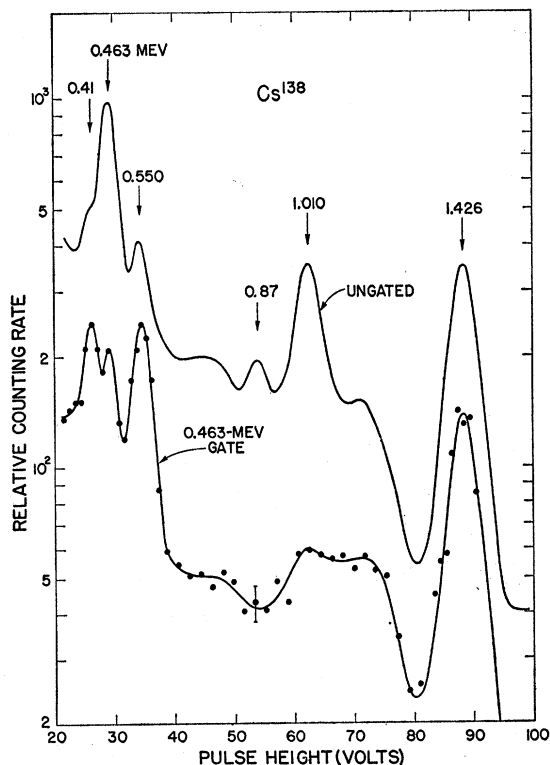


FIG. 3. Gamma-gamma coincidence results. Lower curve: spectrum in coincidence with the 0.43- to 0.52-Mev region. Upper curve: ungated spectrum.

⁵ Bunker, Mize, and Starner, Phys. Rev. **94**, 1694 (1954).

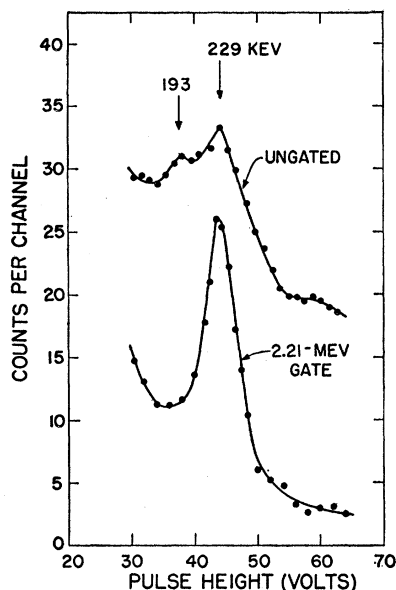


FIG. 4. Gamma-gamma coincidence results. Lower curve: spectrum in coincidence with the 2.21-Mev gamma ray. Upper curve: ungated spectrum.

particles. These measurements were not very informative because of the many beta transitions of not very different energy which appear to be involved in the decay. The gamma-ray spectrum in coincidence with selected portions of the total beta spectrum was also examined. The semiquantitative information which was obtained supported the proposed decay scheme and is discussed in a later section.

Conversion Electron Spectrum

The internal-conversion electron spectrum was examined in a conventional photographic-recording 141-gauss permanent-magnet spectrograph with $\sim 0.25\%$ resolution. The source was deposited on a 10-mil wire by the procedure described above. Because of the short half-life of Cs^{138} and the rather low intensities of the low-energy transitions, most of the lines observed on the photographic plate were rather faint. Table II summarizes the data on the lines which were recorded.

The electron line at 76.0 keV could come either from the K -shell conversion of a 113.4-keV gamma ray following Cs^{138} beta decay, or from the K -shell conversion of a 91.2-keV gamma ray following the beta decay of Rb^{89} . As stated above, the sources used in the permanent magnet spectrograph contained some Rb^{88} and Rb^{89} as impurities, and not enough runs were made to establish the half-life of this line. A careful examination of the 70- to 130-keV region with a gamma-ray scintillation spectrometer did not give conclusive results regarding the existence of a weak 91.2- or 113.4-keV gamma ray. Therefore, a 113.4-keV transition is not included in the decay scheme given in Fig. 5, and we

leave the assignment of the 76.0-keV electron line uncertain.

The other conversion-electron lines correspond quite well to gamma-ray transitions observed with the scintillation spectrometer when Cs^{138} sources free of Rb^{89} were used.

The lines observed on the photographic plate had densities too nearly the same as that of the beta background to allow very accurate relative intensity measurements. The K/L_I conversion ratio for the 0.1389-Mev gamma-ray was found to be ≥ 6 . The L_{II} and L_{III} lines associated with this transition were unobserved, and an upper limit of 0.25 can be placed on the ratio of the intensity of either of these lines to that of the L_I line. The intensities (corrected for spectrograph geometry and film sensitivity) of the L_I line of the 0.1389-Mev transition and the K lines of the 0.1931- and 0.2289-Mev transitions are estimated to be in the ratio 1:1:1.8.

DECAY SCHEME AND DISCUSSION

The level scheme shown in Fig. 5 has been constructed on the basis of the data in Tables I and II. The arguments are as follows:

(1) The 2.63-Mev gamma ray was not found to be in coincidence with any other gamma rays and consequently is taken to be a transition to the ground state. The 3.34-Mev gamma ray is also assumed to lead to the ground state though no direct proof, other than energy considerations, was established.

(2) The 1.426-Mev gamma ray was observed to be in coincidence with the 1.010-Mev gamma ray and with all the members of the complex of gamma rays which is shown in Fig. 5 to have the 1.010-Mev gamma ray as a crossover. The possibility that this arrangement could be inverted, i.e., that the above complex of levels could end on the ground state and be preceded by the 1.426-Mev gamma ray, is eliminated by the much higher intensity of the 1.426-Mev gamma ray. In addition, such a possibility would be inconsistent with the gamma-beta coincidence experiments. In the gamma-ray spectrum coincident with the portion of the beta spectrum above 2.95 Mev, the ratio of the peak height of the 1.010-Mev gamma ray to that of the 1.426-Mev gamma ray was observed to be 2:9. When the gamma

TABLE II. Internal conversion electron energies observed with a permanent magnet spectrograph.

Energy of electron line (keV) ^a	Conversion shell assumed	Corresponding gamma transition (keV)
76.0	...	see text for assignment
101.5	K	138.9
133.2	L_I	139.2
155.7	K	193.1
191.5	K	228.9
373.2	K	410.6
425.2	K	462.6

^a The uncertainty in energy is ± 0.3 keV.

spectrum was instead gated with the lower-energy half of the beta spectrum, this ratio was found to be 2:3. This proves that the 1.426-Mev gamma ray follows the 1.010-Mev gamma ray. Finally, from the systematic variation of the energy of the first excited states of even-even nuclei as a function of neutron number,⁶ it is most unlikely that the first excited state of Ba¹³⁸ could be at either 0.139 or 0.463 Mev; however, a first excited state at 1.426 Mev agrees very well with the systematics.

(3) The complex of gamma rays discussed in (2) which precedes the 1.426-Mev gamma ray can only be arranged as drawn, or in the sequence obtained by inverting the whole group, in order to be in accord with the energy and coincidence data. The inverted order is not possible for intensity reasons (the very intense 0.463-Mev gamma ray must be at the bottom) and also because of the way in which the gamma-ray coincidence spectrum was observed to change when gated by different selected portions of the beta spectrum.

(4) The coincidence measurements showed that the 2.21- and 0.23-Mev gamma rays are in cascade (see Fig. 4) but did not establish the order of emission. However, the fact that the 2.21-Mev gamma is by far the more intense provides a strong argument for postulating that it must follow the 0.23-Mev gamma. Since the sum of the energies of these two gammas is 2.44 Mev, it is assumed that this cascade parallels the (1.010-Mev,

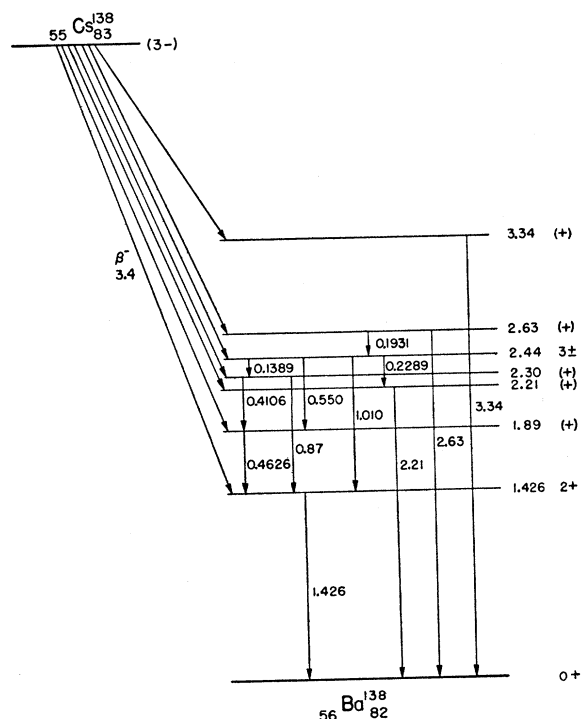


FIG. 5. Proposed decay scheme of Cs¹³⁸. Energies are given in Mev.

⁶ G. Scharff-Goldhaber, Phys. Rev. 90, 587 (1953); P. Preiswerk and P. Stähelin, Nuovo cimento 10, 1219 (1953).

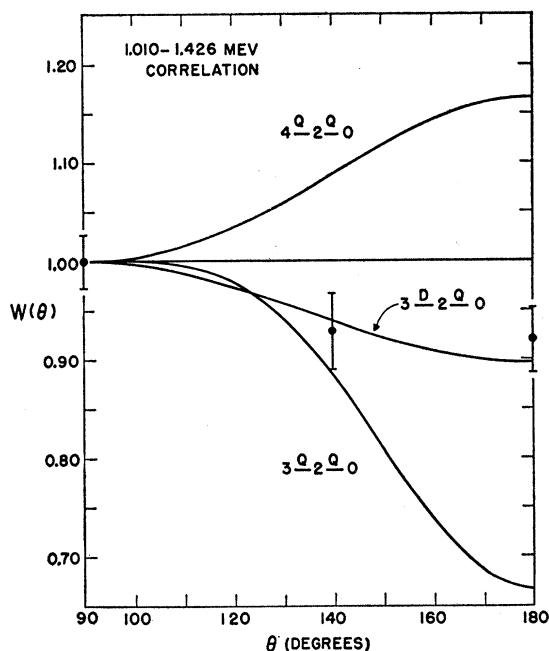


FIG. 6. Angular correlation of the 1.010- and 1.426-Mev gamma rays. The experimental values were normalized by dividing by $W(90^\circ)$. The solid curves are theoretical correlations.

1.426-Mev) cascade. It should be noted that the measured value of the number of 1.426-Mev gammas per disintegration provides an additional proof that both the 2.21- and 2.63-Mev gamma rays are ground-state transitions.

(5) The position of the 0.193-Mev gamma ray was not verified by coincidence experiments, but the energy agreement in the position shown is very good.

(6) The gamma ray which gives rise to the 0.076-Mev internal-conversion electrons does not appear in the decay scheme since it was not established that this gamma belongs to Cs¹³⁸.

The sequence of levels in Ba¹³⁸ is therefore established quite definitely by the above arguments. There is not enough information available to establish the spins of all of the levels, but in order to obtain some information on this point a measurement of the angular correlation of the (1.010-Mev, 1.426-Mev) gamma-ray cascade was made. Since this cascade is the only one in the decay scheme in which both gamma rays have energies greater than 1 Mev, the angular correlation measurement could be made unambiguously, though with rather poor statistics because of the short half-life.

The directional correlation pattern was studied by measuring the coincidence counting rate as a function of the angle subtended at the source by two NaI(Tl) detectors. The NaI(Tl) crystals, both 2×2-inch cylinders, were placed in lead collimators and shielded from direct beta rays with $\frac{3}{4}$ inch of Lucite. In addition, a 0.1-inch thick lead absorber was used in front of each crystal to attenuate backscattered radiation. The full

TABLE III. Beta branching of Cs¹³⁸ calculated from the measured gamma-ray intensities of Table I.

Beta group end point (Mev)	Percent	log ft
3.4	21	7.59
2.94	12	7.60
2.62	16	7.25
2.53	4.5	7.74
2.39	36	6.70
2.20	10	7.11
1.49	0.5	7.73

width at half-maximum of the angular resolution curve was measured to be 11.0°. The Cs¹³⁸ sources, which consisted of ~50 microliters of active solution, were contained in a $\frac{1}{8}$ -inch diameter cylindrical cavity in the end of a $\frac{1}{4}$ -inch diameter Lucite rod.

Conventional electronic circuits were used. A fast ($2\tau=3.6\times 10^{-8}$ sec) coincidence requirement was imposed on the unselected pulses; subsequently, a 0.4- μ sec triple coincidence requirement was imposed on the output pulses of the fast coincidence circuit and of the two single-channel analyzers which provided the necessary energy discrimination. The upper and lower energy bounds of both single-channel analyzers were set at 1.65 and 0.95 Mev, respectively.

The results of the measurement are shown in Fig. 6, where the experimental points have been plotted along with the theoretical correlation functions for the cascades 3(D)2(Q)0, 3(Q)2(Q)0, and 4(Q)2(Q)0. The reason for choosing these particular correlations for comparison is that if one makes the very reasonable assumption that the 1.426-Mev level is a 2+ state,^{6,7} then from beta and gamma selection rules the 2.44-Mev level must have a spin of 3 or 4. The data are clearly in best agreement with a 3(D)2(Q)0 correlation. Of course a small amount of quadrupole mixing in the first transition cannot be excluded.

From the gamma-ray intensities of Table I, the level scheme of Fig. 5, and the observed beta-ray spectrum end point,¹ one can calculate the energies and intensities of all the beta groups of Cs¹³⁸.⁸ The results of these calculations, along with the corresponding log ft values,

⁷ M. Goldhaber and A. W. Sunyar, Phys. Rev. **83**, 906 (1953).

⁸ The beta spectrum discussed in reference 1 was found to be very complex and its analysis into unique groups was virtually impossible. The measured beta spectrum is approximately, but not exactly, reproduced by addition of the groups specified in Table III.

are shown in Table III. § The ft values suggest that all of the observed excited levels have the same parity (assumed to be positive since the first excited states of even-even nuclei invariably have positive parity^{5,6}). This would imply that none of the observed gamma transitions involve a parity change. On the other hand, the internal conversion data on the 0.1389-Mev gamma ($K/L_I \geq 6$; $L_I:L_{II}:L_{III}=1:<0.25:<0.25$) are consistent with either an $M1$ or $E1$ assignment for this transition.⁹ Therefore, we cannot exclude the possibility that the 2.44-Mev level is a 3- state, although the 2.39-Mev beta transition would then have an anomalously high ft value.

The spin and parity of Cs¹³⁸ most nearly consistent with the above data is 3-. From the standpoint of nuclear shell structure, this state probably has a ($d_{5/2}, f_{7/2}$) configuration. It is noteworthy that La¹⁴⁰, which differs from Cs¹³⁸ by only two protons, also has a spin of 3-.¹⁰ In fact, the entire decay scheme of La¹⁴⁰^{10,11} bears a remarkable similarity to that of Cs¹³⁸. Both daughter nuclides, Ce¹⁴⁰ and Ba¹³⁸, have first excited states occurring at ~1.5 Mev, above which are a number of relatively closely-spaced levels. A fact of interest is that in both level schemes the ratio of the energy of the first excited state (E_1) to that of the second excited state (E_2) is ~1.3, whereas the great majority of even-even nuclides with $36 \leq N \leq 88$ have E_2/E_1 ratios ranging between 2 and 2.5.^{12,13} This considerable deviation from the general trend is probably associated with the closed-neutron-shell structure of Ce¹⁴⁰ and Ba¹³⁸. Support for this hypothesis is found in the fact that most of the other even-even nuclei in the above N region which exhibit anomalously low E_2/E_1 ratios (e.g., Sr⁸⁸,¹⁴ Zr⁹⁰,¹⁵ and Sn¹¹⁶¹⁶) have closed shells or closed subshells of neutrons or protons.

§ Note added in proof.—Evidence for additional low-energy beta groups has been obtained by Dr. N. H. Lazar, of Oak Ridge National Laboratory, who has observed weak gamma rays with energies >3.34 Mev (private communication.)

⁹ Rose, Goertzel, Harr, Spinrad, and Strong, Phys. Rev. **83**, 79 (1951); Rose, Goertzel, and Swift (privately circulated tables).

¹⁰ Bolotin, Pruett, Roggenkamp, and Wilkinson, Phys. Rev. **99**, 62 (1955).

¹¹ G. R. Bishop and J. P. Perez Y Jorba, Phys. Rev. **98**, 89 (1955).

¹² G. Scharff-Goldhaber and J. Weneser, Phys. Rev. **98**, 212 (1955).

¹³ L. Wilets and M. Jean, Phys. Rev. **102**, 788 (1956).

¹⁴ Lazar, Eichler, and O'Kelley, Phys. Rev. **101**, 727 (1956).

¹⁵ K. W. Ford, Phys. Rev. **98**, 1516 (1955).

¹⁶ Slätis, du Toit, and Siegbahn, Phys. Rev. **78**, 498 (1950); Arkiv Fysik **2**, 321 (1950). Stähelin, Maeder, and Pochon, Z. Physik **140**, 498 (1955).