ments at 90° to the incident photon beam. The experimental results for both photofission and fast neutron fission are at least qualitatively compatible with the collective model picture of the fission process[10] whereas they are in disagreement with predictions to be obtained from the simple liquid drop model.

We express our thanks to Dr. J. H. Coon and the Los Alamos Cockcroft-Walton group for making available to us their accelerator and necessary auxiliary equipment required to monitor the neutron beam. We also thank the Los Alamos homogeneous reactor group for the use of their reactor. We are indebted to Dr. Keith Boyer for suggesting the type of collimator used in these experiments and to Dr. D. L. Hill for several stimulating interpretative discussions on the subject of the collective model.

[10] For discussion refer to reference 3, page 1116.

PHYSICAL REVIEW VOLUME 94, NUMBER 3 MAY 1, 1954

Inner Bremsstrahlung of Cs^{131}†

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(Received January 13, 1954)

The continuous gamma-ray spectrum (inner bremsstrahlung), accompanying orbital electron capture in Cs^{131}, has been observed by the method of scintillation spectrometry. The absolute intensity of the gamma radiation has been measured and compared with the total disintegration rate. The end point of the gamma-ray distribution was found to be 320±10 kev. The experimentally determined shape of the spectrum, as well as the intensity of the continuum, is compared with theoretical calculations. The disagreement exceeds considerably that which might be expected from a consideration of the probable errors of the observations. The growth of the 12-day isomer of Xe^{131} was not detected.

INTRODUCTION

THE continuous gamma radiation (inner bremsstrahlung) associated with the electron capture process was first investigated theoretically by Morrison and Schiff.[1] More recently, Jauch[2] has reviewed and extended the earlier studies. In addition to experimental comparisons of the spectral distributions which have been computed theoretically, this particular disintegration process can also provide information concerning the disintegration energy released in electron capture.

The gamma-ray continuum has been previously detected,[3–7] and the spectral distributions determined for the cases of Fe^{55}, A^{37}, and Ge^{71}. However, it appears that the extent of gamma-ray emission per disintegration has not been measured for any of the above-cited examples. The radioactive chain $Ba^{131} \rightarrow Cs^{131} \rightarrow Xe^{131}$ was first reported by Yu, Gideon, and Kurbatov.[8] The results of several subsequent investigations showed that neither gamma rays nor conversion electrons are emitted.[9–11] Cheng and Kurbatov[12] have reported the growth of the 12-day Xe^{131m} from Cs^{131}, whereas Canada and Mitchell[13] obtained a contrary result. To study the properties of the inner bremsstrahlung and to investigate further the possible formation of Xe^{131m}, the radiations of Cs^{131} have been reinvestigated.

CHEMICAL PROCEDURE

Eighty-eight grams of $Ba(NO_3)_2$ were exposed to neutrons in the Oak Ridge pile for a period of four weeks. From an aqueous solution of this $Ba(NO_3)_2$ with its cesium daughter element, $BaCl_2 \cdot H_2O$ was precipitated by ether and HCl, thus removing barium alone. Small portions of inactive $Ba(NO_3)_2$ were repeatedly added and precipitated as $BaCl_2 \cdot H_2O$ to extract all of the active barium from the cesium solution. The cesium solution was evaporated to small volume and again scavenged by $BaCl_2 \cdot H_2O$ precipitation. Finally, after adding approximately one milligram of cesium carrier—$CsClO_4$ was precipitated.

The first separation of cesium from the neutron irradiated barium disclosed the presence of a 660-kev gamma ray in the cesium fraction which decayed with a half-period of approximately seven days and may, therefore, be associated[14] with Cs^{132}.

† Assisted by the joint program of the U. S. Office of Naval Research and the U. S. Atomic Energy Commission.
* Research Fellow, Bartol; on leave of absence from Agra College, Agra, India.
[1] P. Morrison and L. I. Schiff, Phys. Rev. **58**, 24 (1940).
[2] J. M. Jauch, Oak Ridge National Laboratory Report ORNL-1102, 1951 (unpublished).
[3] H. Bradt *et al.*, Helv. Phys. Acta **19**, 222 (1946).
[4] Bell, Jauch, and Cassidy, Science **115**, 12 (1952).
[5] D. Maeder and P. Preiswerk, Phys. Rev. **84**, 595 (1951).
[6] C. A. Anderson and G. W. Wheeler, Phys. Rev. **90**, 606 (1953).
[7] Saraf, Varma, and Mandeville, Phys. Rev. **91**, 1216 (1953).
[8] Yu, Gideon, and Kurbatov, Phys. Rev. **71**, 382 (1947).
[9] S. Kotcoff, Phys. Rev. **72**, 1160 (1947).
[10] E. Kondaiah, Arkiv Fysik **2**, 295 (1951).
[11] R. Canada and A. C. G. Mitchell, Phys. Rev. **83**, 76 (1951).
[12] L. S. Cheng and J. D. Kurbatov, Phys. Rev. **78**, 319 (1950).
[13] R. Canada and A. C. G. Mitchell, Phys. Rev. **81**, 382 (1947).
[14] L. M. Langer and G. Ford (unpublished).

In order to remove all traces of Cs^{132}, about 20 mg of cesium was added to the barium solution and precipitated as the perchlorate. Cs^{131} was then allowed to grow from the Ba^{131}. After 10 days the barium was precipitated as $BaCl_2 \cdot H_2O$ and pure carrier-free Cs^{131} was obtained from the supernatant liquid by evaporation with H_2SO_4. The residue was heated with $(NH_4)_2CO_3$ to remove excess H_2SO_4. The source was prepared by evaporation of $CsSO_4$ solution on a thin Formvar film over a circular area of 5-mm diameter.

EXPERIMENTAL PROCEDURE

In Fig. 1 is shown the experimental arrangement for measurement of the gamma-ray spectrum. The crystal detector of NaI-Tl was cylindrical in shape, 3.5 cm in diameter and 3.5 cm in height. Because of the low intensity of the effect, it was not possible to collimate the beam of gamma rays by the introduction of a slit system or by increasing the distance between the source and the crystal. In the present investigation, the distance between the source and the top of the crystal was 1.25 cm. The scintillation spectrometer was calibrated by gamma rays of energies 70, 157, and 209 kev emitted from Au^{199}, 280 kev from Hg^{203}, 320 kev from Cr^{51}, and 662 kev from Cs^{137}. The pulse heights were found to be linear with energy over the above-indicated region of energy. The energy resolution (full width of the photopeak at half-maximum) for the 662-kev gamma ray of Cs^{137} was eight percent. The intensity of the inner bremsstrahlung is several orders of magnitude less than that of the 30-kev x-rays of Xe^{131} which are emitted after K capture in Cs^{131}. With the source in close proximity to the crystal, there was the strong possibility of "pile up" of the voltage pulses associated with the intense x-rays. This x-ray intensity was suppressed by placing copper absorbers between the source and the sodium iodide crystal. As shown in Fig. 2, the gamma-ray spectrum was recorded through several different thicknesses of the x-ray absorbing copper. A channel width of 1 volt (13.3 kev) was used. Below 150 kev counts were recorded at energy intervals of one-half volt.

To determine the probability per disintegration of emission of a gamma ray of the continuum, it was

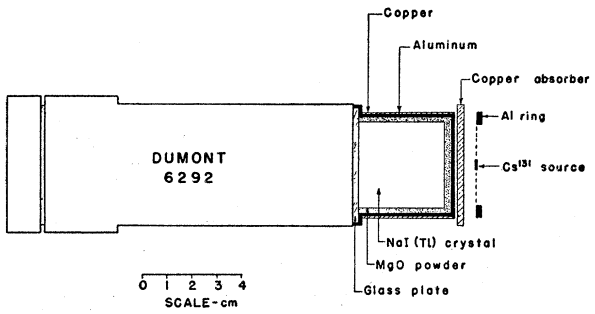

FIG. 1. Experimental arrangement for detection of the continuous gamma-ray spectrum.

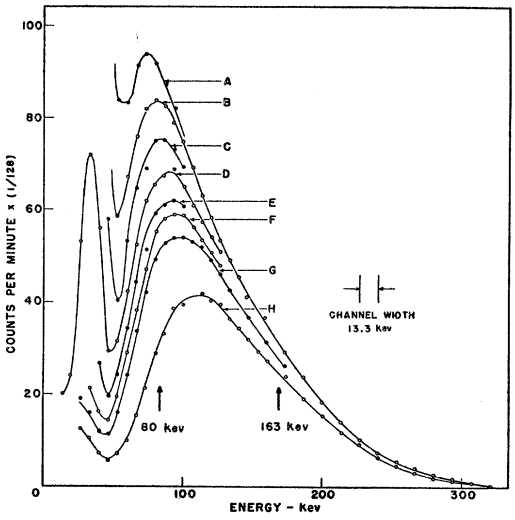

FIG. 2. Gamma-ray spectrum with various intervening amounts of copper absorber: $A=710$ mg/cm^2; $B=840$ mg/cm^2; $C=970$ mg/cm^2; $D=1100$ mg/cm^2; $E=1230$ mg/cm^2; $F=1360$ mg/cm^2; $G=1490$ mg/cm^2; $H=2200$ mg/cm^2.

necessary to have an estimate of the strength of the source of Cs^{131}. To obtain an estimate of the number of disintegrations per second occurring in the source, the $CsSO_4$ previously used for observation of the quantum continuum was dissolved in 5 cc of H_2O. Of this solution, 0.1 cc was further diluted to 5-cc volume, and 0.02 cc of this amount was dried over a thin Formvar film to cover an area of 0.5 cm^2. This source was mounted upon a second crystal of NaI-Tl, 1.5 cm in diameter, and of thickness 0.64 cm. The amount of absorber intervening between source and crystal was 3 mg/cm^2 of aluminum. Pulses of such height as to correspond to a quantum energy of 30 kev were counted. In computing the absolute intensity of the x-radiation and hence the source strength, the solid angle was estimated to be fifty percent, and the intrinsic efficiency of detection in the crystal was taken to be one-hundred percent. Because the results of the measurement of the fluorescence yield of xenon have been at variance, the theoretical value,[15] calculated nonrelativistically, has been used.

The copper for absorption of the x-radiation is, of course, also effective in absorbing the continuum of gamma rays under study. In addition to this absorption, Compton scattering in the copper may result in the appearance at low energies of a contribution from regions of the spectrum of much higher energy. Similarly, Compton scattering in the crystal itself and subsequent escape of the recoil photons can also distort the spectral distribution.

In order to ascertain the extent of these several effects, the absorption curves of Fig. 3 were obtained from the data of Fig. 2. The counting rate in a channel

[15] E. H. S. Burhop, *Auger Effect* (Cambridge University Press, Cambridge, 1952), p. 45.

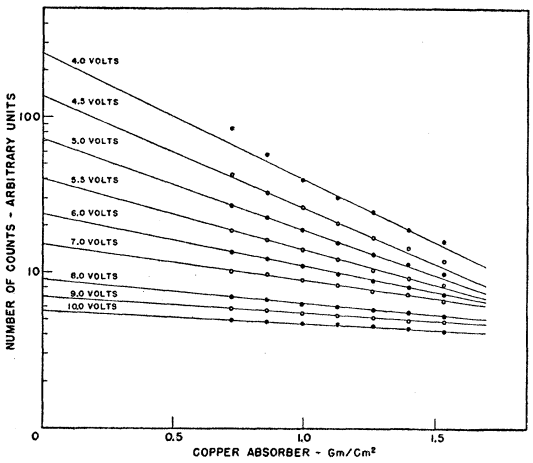

FIG. 3. Some of the data of Fig. 2 plotted as a function of copper absorber thickness at given quantum energies along the continum. The corresponding energy values are 4.0 volts—54 kev, 4.5 volts—60.5 kev, 5.0 volts—67 kev, 5.5 volts—73.5 kev, 6.0 volts—80 kev, 7.0 volts—94 kev.

width of one volt at various different pulse-height settings is plotted as a function of copper absorber thickness. From the apparent linearity of the absorption curves on the semilogarithmic plot, it is concluded that the aforementioned effects are small. Finally, extrapolation of the curves to zero absorber thickness removes any nonuniformities introduced by the variation along the spectrum of the absorption coefficient of copper.

To show further that the counting rate at each pulse height arose primarily from the total absorption in the crystal of gamma radiation of the proper corresponding energy, the half-value thickness of the counting rates at several different pulse heights of Fig. 3 are shown in Table I. It is to be noted that these comparisons are confined to the region of lower energies, where Compton scattering contributions from gamma rays of higher energy are larger than in any other part of the spectrum. The observed half-value thicknesses are compared with those for monoenergetic radiations as given by Compton and Allison.[16] The relatively good agreement between the observed and expected values of the half-thicknesses shows that any distortion of the low-energy region of the spectrum by the Compton effect at higher energies is at most a few percent.

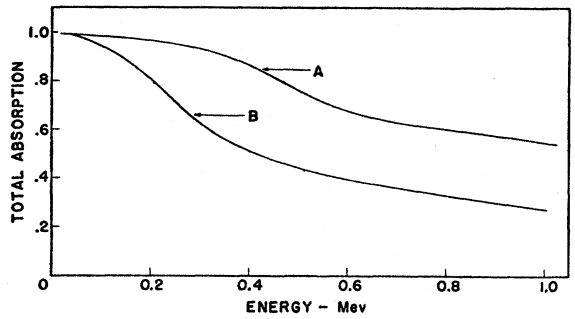

FIG. 4. Efficiency of detection as a function of energy calculated from the total absorption coefficient. Curve *A*, for gamma radiation at normal incidence relative to the face of the crystal. Curve *B*, source distance 1.25 cm from crystal surface.

In order to evaluate the counting rates at the pulse heights corresponding to low energies as shown in Fig. 3, the counting rates were extrapolated to zero absorber thickness at all points of the spectrum. The experimentally measured spectrum thus obtained is compared, after making a suitable resolution correction with the theoretically expected distribution. Before making the final comparison, however, the theoretical distribution was altered to take into account (1) variation of detection efficiency with quantum energy; (2) distortion arising from contributions to the spectrum of the Compton effect; and (3) escape, from the crystal of the x-rays of iodine.

THE CORRECTIONS

The experimental curve obtained by the extrapolation process is corrected for finite resolution by applying the following expression:[17]

$$N_c(E)=N_e(E)-KN_e'(E)-\tfrac{1}{2}KEN''(E),$$

where $N_c(E)$ is the corrected energy distribution, and $N_e(E)$ is the experimentally observed distribution. $N'(E)$ and $N''(E)$ are the first and second derivatives of $N_e(E)$; $K=[W(E)]^2/(0.693\times 2E)$, where $W(E)$ is the half-width at half-maximum of a photopeak produced by radiation of energy E.

TABLE I. Half-value thicknesses of the counting rates for several different pulse heights.

Pulse height—volts	4.0	4.5	5.0	5.5	6.0	7.0
Channel width—volts	1.0	1.0	1.0	1.0	1.0	1.0
Energy—kev	54	60.5	67	73.5	80	94
Half-thickness in Cu—mg/cm² (Compton and Allison)	325	430	540	655	900	1300
Half-thickness in Cu—mg/cm² (These measurements)	290	400	520	630	850	1200

As stated in the previous section, the theoretical distribution was subjected to three corrections before comparison with experiment. These corrections are:

(1) Correction for Variation of Detection Efficiency with Quantum Energy

Were the source at an infinite distance from the crystal, so that normal incidence would occur, the efficiency of detection for a given energy would depend solely upon the absorption coefficient[18] and the thickness of the crystal. This efficiency *versus* quantum energy for the crystal used in these investigations is

[16] A. H. Compton and S. K. Allison, *X-rays in Theory and Experiment* (D. Van Nostrand Company, Inc., New York, 1935).
[17] T. B. Novey, Phys. Rev. **89**, 672 (1953).
[18] W. H. Jordon, Ann. Rev. Nuc. Sci. **2**, 221 (1952).

shown in Fig. 4, curve A. This curve is to be compared with curve B where the actual source distance of 1.25 cm is taken into account. In calculating curve B, it was assumed, as is the case, that the entire face of the crystal is under irradiation.

(2) Correction for Presence of Compton Effect

When monochromatic radiation is incident upon a crystal of NaI-Tl, a peak of pulses appears which corresponds to a loss of all the quantum energy in the crystal. Because of the escape of the recoil photon, the Compton effect leads to a distribution of pulse heights, extending from zero to a certain maximum value. Figure 5 shows the percentage of all the pulses generated in the NaI-Tl crystal which falls in the peak corresponding to total absorption of the quantum energy. Curve B is a plot of the ratio of the photoelectric absorption coefficient to the total absorption coefficient. Curve A is the measured ratio of the counts in the peak corresponding to total energy loss to the total number of counts. Curve A was obtained from observations on a series of monochromatic gamma-ray sources.

(3) Correction for Escape of the X-Radiation of Iodine

Because of the rapid absorption of the softer gamma rays of the continuum in NaI(Tl), the probability of escape of the associated x-rays of iodine is greater at the lower spectral energies; that is, since the soft gamma rays of the continuum are almost totally absorbed in a thin layer of NaI(Tl) at the surface of the crystal, the chance of escape of the x-rays from the same thin layer is great. At 33.7 kev, the K-absorption edge of iodine, the escape probability is at a maximum. This escape correction has been calculated by Novey.[17]

The theoretical distribution is plotted as curve A in Fig. 6 according to the equation[1,2]

$$\frac{dP}{P_0}=\frac{\alpha}{\pi(mc^2)^2}\left(1-\frac{E}{E_0}\right)^2 EdE,$$

where P_0 is the total number of disintegrations per unit time, dP the probability of photon emission of energy between E and $E+dE$, and E_0 the end point of the gamma-ray spectrum. The electron rest mass is denoted by m, and α is the fine structure constant 1/137. Curve B is a plot of curve A corrected for variation with energy of the detection efficiency, paragraph (1) above. Curves C and D show how the counts of curve B must be redistributed between total-energy absorption on the one hand, and Compton effect and x-ray escape on the other, curve D being the sum of the contributions at lower energies arising from Compton effect and the x-ray escape process. Curve E, the sum of curves C and D, is the expected experimental curve, if the initially assumed theoretical distribution is correct.

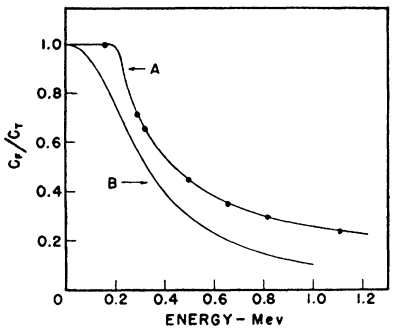

FIG. 5. Curve A—Fraction of pulses as a function of energy corresponding to absorption of the photons' full energy; C_F/C_T experimentally determined. Curve B—Ratio of photoelectric absorption coefficient to total absorption coefficient for NaI as a function of quantum energy.

SEARCH FOR MONOENERGIC GAMMA RAYS FROM Cs^{131}

It is evident that were any monoenergic nuclear gamma rays present in the decay of Cs^{131}, they would severely distort the continuous spectrum, or if having any appreciable intensity, might completely overwhelm the faint radiation of the continuum. The measured distribution of gamma rays from Cs^{131} shown in Fig. 2 gives an end point at 320±10 kev. From a consideration of the decay scheme[19] of $I^{131} \xrightarrow{\beta} Xe^{131}$, it appears that this disintegration energy is sufficient to excite levels in Xe^{131} which on deexcitation would emit gamma rays of energies 163 kev and 80 kev.

Althouth no real evidence for the presence of monochromatic radiation of energy 80 kev appears on the curves of Fig. 2, further measurements were performed to place an upper limit on its intensity. The radiation of the continuum was absorbed in platinum and gold as

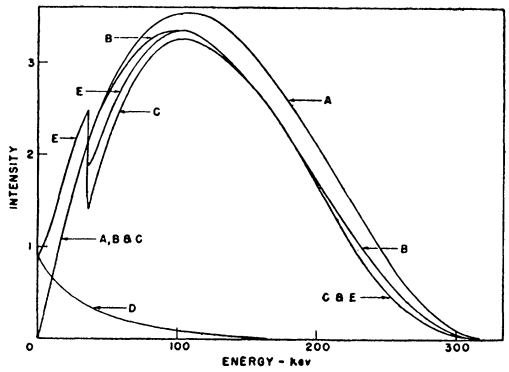

FIG. 6. Calculated energy distribution. A—Theoretical spectrum. B—Modified for detection efficiency. C—Distribution of pulses corresponding to absorption of full energy of the photon. D—Counting rate arising from escape of Compton recoil photons and iodine x-rays. E—Sum of contributions of curves C and D, the expected distribution calculated from theory and altered to take into account experimental conditions.

[19] M. Goldhaber and R. D. Hill, Revs. Modern Phys. 24, 216 (1952).

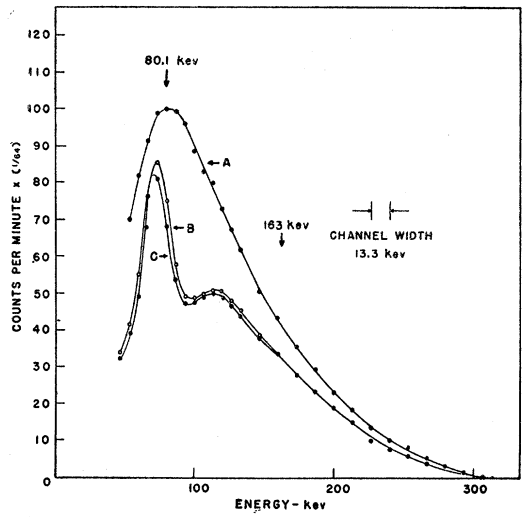

FIG. 7. Absorption of the continuous spectrum in copper, copper and gold, and copper and platinum. Curve A—860 mg/cm² Cu. Curve B—860 mg/cm² Cu+107 mg/cm² Au. Curve C—860 mg/cm² Cu+97 mg/cm² Pt. Gold and platinum foils were placed above the copper which was immediately over the canned crystal.

shown in Fig. 7. The K-absorption edge of Au occurs at 80.5 kev and that of Pt at 78.1 kev. The energy of the sought for gamma ray is accurately known by crystal spectrometer measurements[20] to be 80.133 Kev; therefore, the radiation would be more heavily absorbed in Pt than in Au. In particular, in the case of the experiments of Fig. 7, the counting rate at 80 kev would have been reduced by approximately 16 percent in the case of Au and 50 percent for Pt. Examination of the curves of Fig. 7 shows this not to be the case. The absorption in Au is slightly less than that in Pt, because the gold foil (97.5 mg/cm²) was somewhat thinner than the platinum one (107.5 mg/cm²). When this difference in thickness is taken into account, the two absorption curves are identical. From these observations, it was estimated that the electron capture branch terminating at the 80-kev level must have an incidence of less than 10^{-7} per disintegration.

In order to place an upper limit upon the branch which might lead to the 163-kev isomeric level of half-period twelve days, a separate experiment was performed. One gram of active $BaCl_2$ was enclosed in a glass bulb and connected to a similar empty glass bulb by an inverted "U" tube. The entire system of two bulbs and connecting tube was then filled with argon to a pressure of one-half atmosphere. After twenty days, the empty bulb was cooled with liquid air and the $BaCl_2$ was heated to 100°C. This process was repeated several times to insure transfer to the empty bulb of any Xe grown from Ba-Cs. The bulb containing any Xe was then sealed off and examined with the scintillation spectrometer. No evidence of a gamma ray at 163 kev was obtained. Taking into account the source-strength, the time of growth, the detection efficiency, and the conversion coefficient of the 163-kev gamma ray, the upper limit for formation of the 12-day metastable state was estimated to be 10^{-10} per disintegration.

[20] Lind, Brown, Klein, Muller, and Dumond, Phys. Rev. **75**, 1544 (1949).

DISCUSSION OF RESULTS

The end point of the continuous gamma-ray spectrum of Cs^{131} occurs at 320±10 kev; therefore, the disintegration energy of Cs^{131} or the mass difference of Cs^{131} and Xe^{131} is 353±10 kev (taking the binding energy of K electron to be 33 kev).

According to the shell model of the nucleus, the orbital of the ground state of Cs^{131} is $d_{\frac{5}{2}}$. This value has been observed in a recent experimental measurement.[21] The residual nucleus, Xe^{131}, is also the decay product of the 8-day I^{131}. From a study of the gamma rays of I^{131}, orbitals of the first two excited states of Xe^{131} have been obtained.[19] The ground state orbital of Xe^{131} has been measured.[19] The spin and parity assignments are summarized in Fig. 8.

Log ft for the electron-capture transition terminating at the ground state of Xe^{131} has been calculated from the currently reported energy measurement and is 5.3. In terms of the two measured ground-state orbitals, the transition may be described as having the selection rules $d_{\frac{5}{2}} \rightarrow d_{\frac{3}{2}}$, $\Delta I = 1$; No. These are the properties of an allowed transition if G-T selection rules are followed. If classified according to Mayer, Moszkowski, and Nordheim,[22] the measured value of log ft does indeed correspond to an allowed transition.

Transitions leading to the 80-kev and 163-kev levels would produce changes in quantum numbers given by $d_{\frac{5}{2}} \rightarrow s_{\frac{1}{2}}$, $\Delta I = 2$; No, and $d_{\frac{5}{2}} \rightarrow h_{11/2}$, $\Delta I = 3$; Yes, respectively, which transitions would, according to G-T rules, be second and third forbidden. Values of log ft calculated from upper limits of the intensities of these transitions are about 12 and 15, corresponding[21] to the respective degrees of forbiddance indicated by the selection rules.

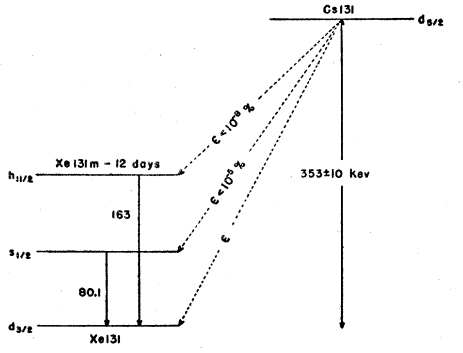

FIG. 8. Disintegration scheme of Cs^{131}.

[21] E. H. Ballamy and K. F. Smith, Phil. Mag. 44, 33 (1953).
[22] Mayer, Moszkowski, and Nordheim, Revs. Modern Phys. **23**, 317 (1951).

The experimentally observed probability of emission of the inner bremsstrahlung is shown in Fig. 9 where it is compared with the theoretically expected result (see curve *E*, Fig. 6). The emission propability per disintegration is plotted in either case; the experimental points are determined as in Fig. 3 by extrapolation of the counting rate at each pulse-height setting to zero copper absorber thickness. Because of various experimental uncertainties in source strength determination, etc., the ordinates of the experimental curve are thought to be in error by as much as fifteen percent. Below 150 kev, because of uncertainties in extrapolation of the counting rates to zero copper absorber thickness, an additional experimental error of perhaps ten percent is thought to be present.

From Fig. 9, it is evident that below 150 kev the discrepancy between experiment and theory becomes too large to be explained by even the liberal estimates of error given above. Because of this sharp disagreement, the question of the purity of the source arises. Evidence for source purity may be cited as follows:

(1) The ratio of the intensity of gamma rays to that of x-rays remained constant over four half-periods of decay of Cs^{131}.

(2) The gamma-ray continuum followed the chemistry of cesium.

(3) The cesium sources were prepared by "milking" barium repeatedly. Each freshly prepared source of cesium exhibited the same gamma-ray continuum.

It should, perhaps, also be mentioned that when a source of cesium was mounted adjacent to a crystal with only 3 mg/cm^2 of aluminum intervening, no charged particles (conversion electrons or beta rays) could be detected. A similar search with a thin-walled Geiger counter likewise yielded no effect.

It is, at the moment, difficult to postulate any definite explanation for the disagreement between experiment and theory. The early theoretical considerations of Morrison and Schiff[1] did not include the effects of the *K*-shell binding energy. The calculations applied only to "low *Z*" nuclei. The effects of possible *L* capture were also neglected. In a private communication, Jauch has given the correction factor for the effect of binding energy to be

$$C=\frac{1+(1+\delta)^2}{2(1+\delta)^2},$$

where δ=binding energy/quantum energy. However, this factor is found to decrease the probability of emission at lower energies and, therefore, enhance rather than decrease the extent of disagreement between

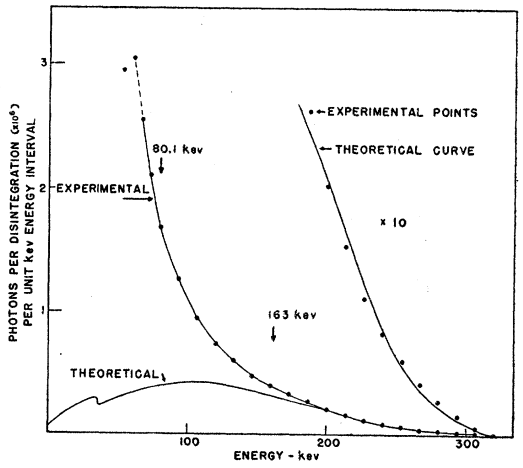

FIG. 9. Photon emission probability per disintegration per unit energy interval (1 kev). The broken portion of the experimental curve is uncertain.

experiment and theory. According to a formula given by Marshak,[23] L_{I} capture occurs in 13.6 percent of the disintegrations and L_{II} capture is one percent. The gamma-ray emission probability associated with *L*-electron capture has not been studied theoretically in great detail.‡

Bremsstrahlung effects relating to the magnetic moment of the emitted neutrino have recently been discussed.[24,25] However, the order of magnitude of the effect has not been accurately estimated.

ACKNOWLEDGMENT

The writer wishes to express his appreciation to Dr. C. E. Mandeville for having originally suggested this research problem and for lending encouragement. He also wishes to acknowledge the many valuable suggestions of Dr. F. R. Metzger, as well as discussions with Dr. L. Eisenbud. The chemical separations were performed by Dr. Walter B. Keighton of Swarthmore College. Finally, the writer is grateful to The Bartol Research Foundation for making possible his stay in the United States and to the Director, Dr. W. F. G. Swann, for his continued interest.

[23] R. A. Marshak, Phys. Rev. **61**, 431 (1942).

‡ *Note added in proof.*—R. E. Cutkosky (Ph.D. thesis, Carnegie Institute of Technology, May, 1953) has calculated the intensity of the inner bremsstrahlung associated with the capture of the *s* and *p* electrons of the *L* shell. The calculations indicate an intensity which is too small to account for the disagreement with theory found in the present measurements.

[24] P. M. Endt, Report of Birmingham Conference on Nuclear Physics, page 29, July, 1953.

[25] J. Weneser, Phys. Rev. **91**, 1025 (1953).