Radiations from Ba^{133}†

BERND CRASEMANN, J. G. PENGRA, AND I. E. LINDSTROM
Department of Physics, University of Oregon, Eugene, Oregon
(Received August 5, 1957)

The decay of the isotope Ba^{133}, produced by pile neutron irradiation of Ba^{132}, has been studied with a magnetic lens and scintillation spectrometers. Gamma rays of 0.079, 0.302, and 0.355 Mev have been measured, with relative intensities 1.00:0.71:2.1 and K-conversion coefficients of 1.3 ± 0.5, 0.11 ± 0.07, and 0.07 ± 0.05, respectively. Conversion electrons from weak transitions of energies 0.158 and 0.276 Mev have been observed and assigned to Ba^{133}. Gamma-gamma coincidences have been measured and a decay scheme is proposed. The K/L capture ratio for Ba^{133} is shown to be greater than 1.5.

I. INTRODUCTION

RECENTLY published work[1–3] on the decay of 9.5-year Ba^{133} has been based on scintillation spectrometer measurements with sources of low specific activity and has produced somewhat inconsistent results. In principle, the long half-life of Ba^{133} makes this isotope suitable for spectrometer calibrations, and sources for this purpose are being marketed commercially by several companies. It was, therefore, considered of interest to re-examine the radiations from Ba^{133}, with special emphasis on a magnetic spectrometer measurement of the conversion electron distribution. A source of rather high specific activity was obtained from the Oak Ridge National Laboratory, where it had been produced by neutron irradiation of a barium target enriched in Ba^{132}.

II. SCINTILLATION SPECTRA

A typical scintillation spectrum of the high-energy gamma rays from Ba^{133} is shown in Fig. 1, and a low-

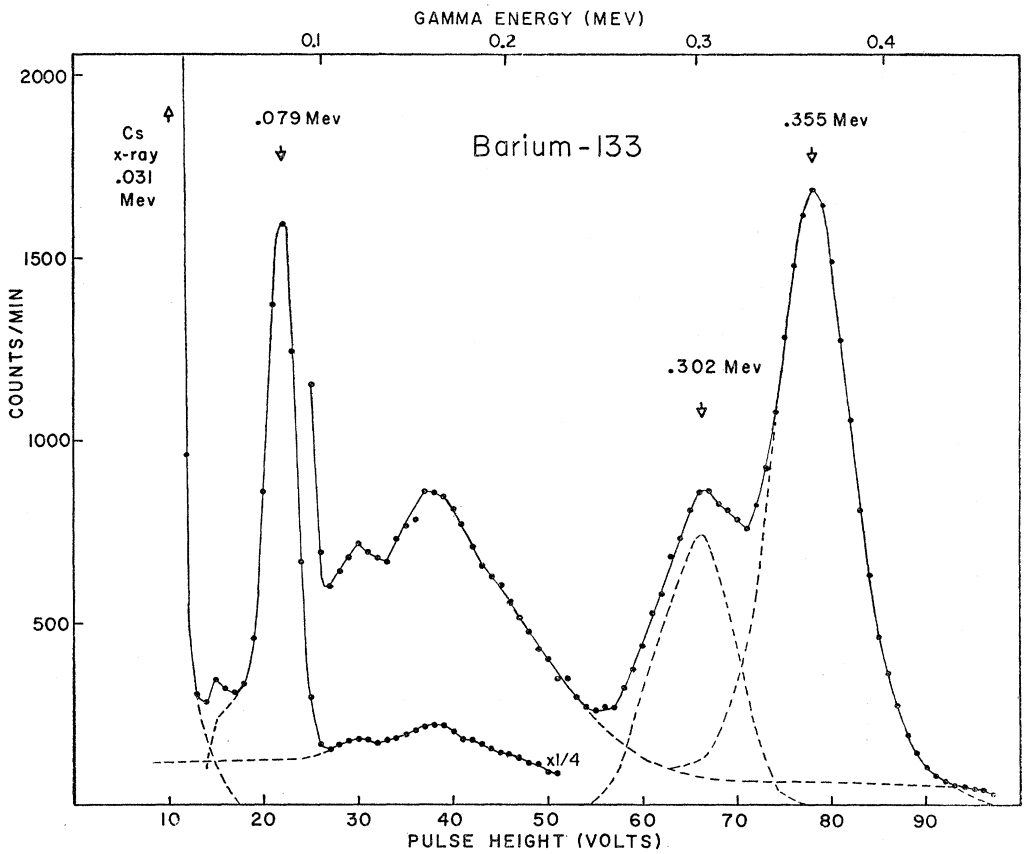

FIG. 1. Scintillation spectrum of Ba^{133} gamma rays, to 0.45 Mev.

† Supported by the National Science Foundation and by the Graduate School of the University of Oregon.
[1] Hayward, Hoppes, and Ernst, Phys. Rev. **93**, 916(A) (1954).
[2] M. Langevin, Compt. rend. **238**, 1310 (1954); **240**, 289 (1955).
[3] M. Langevin, Ann. phys. **1**, 57 (1956).

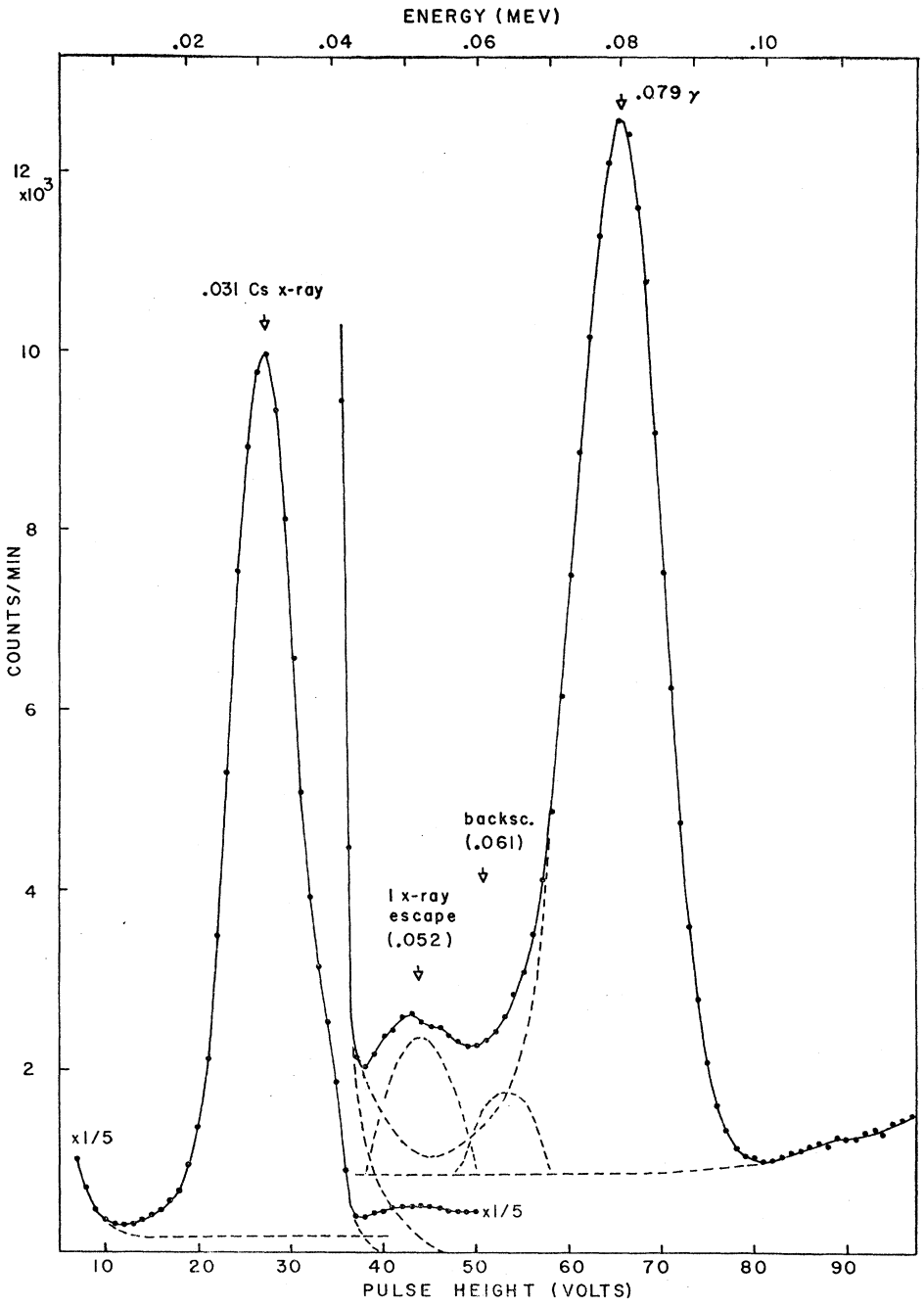

FIG. 2. Scintillation spectrum of Ba^{133} gamma rays, to 0.12 Mev.

energy spectrum is reproduced in Fig. 2. These spectra were obtained with a Harshaw $1\frac{1}{2}\times 1$-in. NaI(Tl) crystal; the source was located centrally below the crystal at a distance of approximately 4 cm. Gamma rays of known energy from Cd^{109} and Se^{75} were used for calibration. The spectra show Ba^{133} gamma rays of approximately 0.079, 0.302, and 0.355 Mev energy. The features appearing between 0.1 and 0.25 Mev in Fig. 1 presumably are due to the Compton distribution and backscatter peaks from the high-energy gamma rays.

In order to compare gamma-ray intensities, the scintillation spectra were analyzed as indicated in Figs. 1 and 2. Corrections for photopeak efficiency were taken from the curves of Kalkstein and Hollander[4] which had been supplemented with experimental points from the Se^{75} spectrum. Results of the intensity comparison are included in Table I.

The low-energy spectrum of Fig. 2 is of special

[4] M. I. Kalkstein and J. M. Hollander, University of California Radiation Laboratory Report UCRL-2764, 1954 (unpublished).

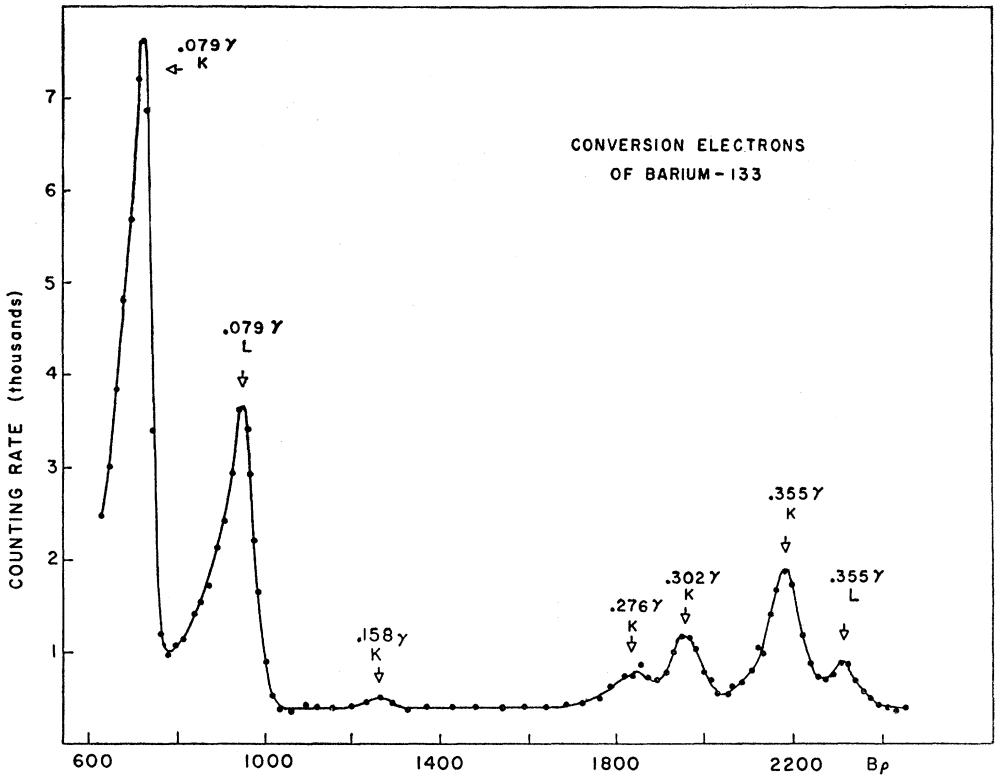

FIG. 3. Conversion electron spectrum of Ba^{133}.

interest, since an 0.057-Mev gamma ray has been reported from scintillation work.[1] If it were of low intensity, such a gamma ray would be difficult to detect, because its energy falls between that of the iodine x-ray escape peak,[5] at approximately 0.051 Mev, and that of a backscatter peak, expected at 0.061 Mev. The area of the x-ray escape peak in Fig. 2 is 7.7% of the total photopeak area, while the theoretical intensity[6] of the escape peak at this energy is 6.5% of the total. The backscatter peak is observed at an energy slightly higher than calculated. This shift is attributed to the occurrence of a rather large number of scattering events on the crystal envelope through angles less than 180 deg. It appears that there is no indication, in the present spectra, of a gamma ray in the 0.050- to 0.060-Mev region, and an upper limit of 5% (as compared with the 0.079-Mev gamma ray) can be set on the intensity of such a ray.

The identification of the feature at 0.052 Mev in Fig. 2 as the iodine x-ray escape peak was further confirmed by comparison with a scintillation spectrum of the 0.087-Mev gamma ray of Cd^{109}, obtained under the same geometry, which shows an escape peak of comparable size.

The identity of features in the 0.05- to 0.06-Mev region of the scintillation spectrum was submitted to a further check by comparing spectra obtained with various thicknesses of platinum absorber between source and crystal. Since the K-absorption edge of platinum is located at 0.07835 Mev, the 0.079-Mev gamma ray will be attenuated more by a platinum absorber than a gamma ray of 0.057 Mev. This difference in attenuation amounts to a factor of 1.7 for an 0.002-in. absorber, and to a factor of 2.7, for an 0.004-in. absorber.[7] If part of the feature around 0.057 Mev were due to an independent gamma ray, then platinum absorbers would make this feature more prominent, as compared with the 0.079-Mev peak. Such an effect was not observed, indicating that the major portion of the spectral distribution between 0.05 and 0.06 Mev is due to secondary processes that involve the 0.079-Mev gamma ray.

Langevin[3] has reported two gamma rays in the 0.080-Mev region, of energies 0.072 and 0.081 Mev. The present scintillation spectra, with and without platinum absorbers, as well as the conversion electron spectrum discussed below, do not resolve more than a single gamma ray in this region. No gamma rays of an energy higher than 0.355 Mev have been observed.

III. CONVERSION ELECTRONS

The conversion electron distribution was measured with a thick magnetic lens spectrometer.[8] The Geiger

[5] P. R. Bell in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland Publishing Company, Amsterdam, 1955), p. 153.
[6] P. R. Bell, reference 5, p. 155.
[7] G. R. White, National Bureau of Standards Report 1003, Washington, D. C., 1952 (unpublished).
[8] B. Crasemann and D. L. Manley, Phys. Rev. **98**, 66 (1955).

TABLE I. Gamma rays from Ba^{133}. (Energies in Mev.)

Transition	Energies from conversion electron spectra: K ce$^-$	L ce$^-$	Gamma	Relative gamma intensity[a]	K/L ce$^-$ ratio	αK	Transition intensity (per decay)	K-shell vacancies (per decay)
Cs K x-ray			0.031	3.8±0.4			1.7[b]	1.7±0.5[b]
γ_1			(0.053)	<0.05				
γ_2	0.0431±0.0005	0.0733±0.001	0.079±0.001	1.00	7.5±0.5	1.3±0.5[c]	1.00	0.52
γ_3			0.079±0.005	<0.15[d]				
γ_4	0.124±0.005		0.158±0.005	<0.2				
γ_5	0.240±0.001		0.276±0.001			high		
γ_6	0.266±0.001		0.302±0.001	0.71±0.07		0.11±0.07[e]	0.26	0.03
γ_7	0.319±0.001	0.350±0.002	0.355±0.002	2.1±0.2		0.07±0.05[e]	0.74	0.05
Ann. rad.			0.511	$<8\times10^{-3}$			$<2\times10^{-3}$[f]	

[a] From scintillation spectra.
[b] Total K vacancies, assuming fluorescence yield 0.87.
[c] By comparison with Cd^{109}.
[d] From coincidence spectra.
[e] By comparison with 0.079-Mev gamma.
[f] Upper limit on decays by positron emission.

tube window was made of 0.67-mg/cm^2 Mylar. The data were corrected for window absorption, using the curves of Saxon,[9] which had been supplemented with experimental points derived from Fermi plots of the electron spectra of S^{35} and Hg^{203}. Sources were prepared by evaporation of a $BaCl_2$ solution in HCl on thin Tygon foils, mounted on Lucite holders.

A typical conversion electron spectrum is shown in Fig. 3. The energies of the observed peaks are listed in Table I. Conversion electrons (ce) from two weak, previously unknown transitions are seen at 0.124 and 0.240 Mev. The corresponding gamma rays, of energies 0.158 and 0.276 Mev, are not apparent in the scintillation spectrum of Fig. 1, where they presumably are masked by other features. The energies of the new transitions fit well into the Ba^{133} decay scheme, which is discussed below. Nevertheless, it was considered necessary to establish that the new transitions do not belong to impurities.

There do not appear to exist any known, long-lived activities which, as contaminants, could produce the 0.124-Mev and 0.240-Mev conversion electrons. Keller and Cork[10] reported 0.2365-Mev conversion electrons from 140-day Ce^{139}, but these electrons were not found subsequently by Ketelle *et al.*[11] There is a 39-hr isomeric state of Ba^{133}, which is known to decay through an 0.276-Mev transition[12]; however, the short lifetime of this isomeric state rules out the possibility that it could be responsible for a conversion electron peak observed here, since the sources used in the present work were over six months old.

In order to confirm that the observed conversion electrons are due to radioactive barium, some of the barium chloride received from Oak Ridge was subjected to additional chemical purification.[13] $BaCl_2\cdot2H_2O$ was precipitated with ether-HCl reagent, redissolved, again precipitated, dissolved, and the solution made basic with carbonate-free ammonia. After scavenging twice with $Fe(OH)_3$, $BaCO_3$ was precipitated, dissolved in 6N HCl, and $BaCl_2$ was again precipitated with ether-HCl reagent. The precipitate was washed repeatedly with absolute alcohol and ether. Sources prepared with this purified barium chloride still showed the 0.124-Mev and 0.240-Mev conversion electrons. It is therefore concluded that these conversion electrons belong to Ba^{133}.

IV. CONVERSION COEFFICIENTS

The K-shell conversion coefficient of the 0.079-Mev gamma ray has been determined by comparison with the conversion coefficient of the 0.087-Mev isomeric gamma ray of Ag^{109}, for which the value 10.3±0.5 has been found by Wapstra.[14] The comparison source was produced by bombarding a silver target with 60 μa-hr of 40-Mev alpha particles in the Crocker Laboratory cyclotron of the University of California. Cadmium was separated from the target by organic solvent extraction, following the procedure of Maxwell, Haymond, Garrison, and Hamilton,[15] and short-lived activities were allowed to decay. The K-conversion electron intensities from a Ba^{133}-source and from a Cd^{109}-source of similar strength were compared in the magnetic lens spectrometer, and the gamma-ray intensities of the two sources were compared in a scintillation spectrometer. From two sets of intensity ratios, obtained with different sources, a value of 1.3±0.5 is computed for the K-shell conversion coefficient of the 0.079-Mev gamma ray of Ba^{133}. Conversion coefficients for the 0.302 and 0.355-Mev gamma rays were obtained by comparison of gamma and conversion electron intensities with those of the 0.079-Mev transition. Results are listed in Table I.

[9] D. Saxon, Phys. Rev. **81**, 639 (1951).
[10] H. B. Keller and J. M. Cork, Phys. Rev. **84**, 1079 (1951).
[11] Ketelle, Thomas, and Brosi, Phys. Rev. **103**, 190 (1956).
[12] R. D. Hill and F. R. Metzger, Phys. Rev. **83**, 455 (1951).
[13] Procedure of Hicks, Folger, and Goeckerman. See W. W. Meinke, University of California Radiation Laboratory Report UCRL-432, 1949 (unpublished).
[14] A. H. Wapstra, Physica (to be published). (Private communication by C. L. McGinnis, Nuclear Data Group, National Research Council.)
[15] W. W. Meinke, reference 13.

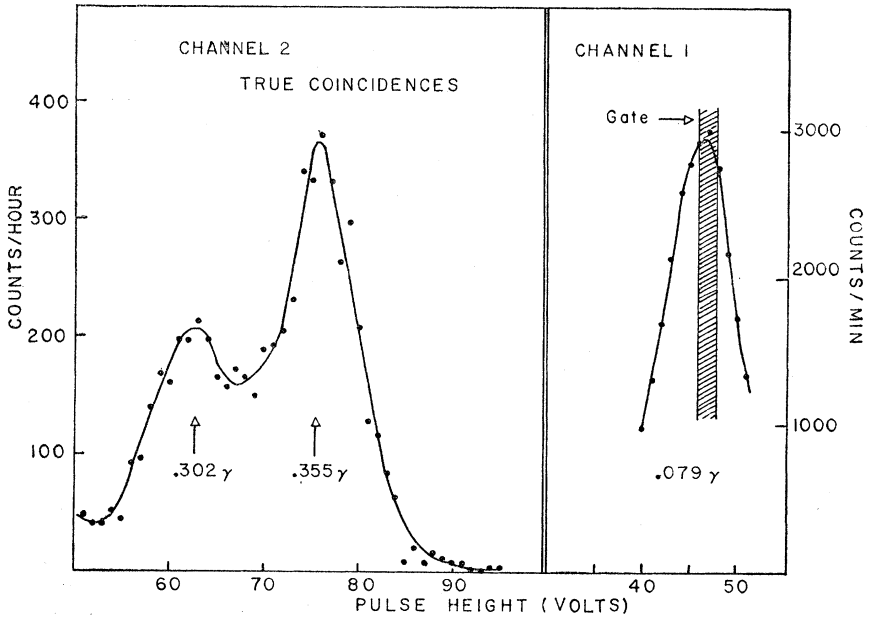

FIG. 4. Ba^{133} gamma-gamma coincidence spectrum.

V. COINCIDENCES

Gamma-gamma coincidences were measured by gating the output of one pulse-height analyzer with the output from the pulse-height analyzer in a second, identical channel. Resolving times of 5 and 2 μsec were employed. The chance coincidence counting rate was determined experimentally by separating the two scintillation counters and repeating each run with two sources, matched in intensity.

Figure 4 shows a section of the coincidence spectrum gated by the 0.079-Mev gamma ray. The spectrum has been corrected for chance counting rate and background. A summary of the coincidence experiments is contained in Table II. The intensity ratio of the 0.355-Mev and 0.302-Mev gamma rays, gated by the Cs K x-ray (2 μsec resolving time) is the same as the intensity ratio of these two gammas in the singles spectrum. This indicates that the mean life of the 0.434-Mev level cannot be much longer than 2 μsec. The (0.079γ) (0.079γ) coincidence rate somewhat exceeds the chance coincidence rate. This result is compatible with the assumption, contained in the proposed decay scheme, that two separate gamma rays of approximately 0.079 Mev occur in the disintegration of Ba^{133}.

TABLE II. Coincidences among radiations from Ba^{133}. (Resolving time 2 μsec.)

Gate \ Coinc.	0.079γ	0.302γ	0.355γ
Cs K x-ray		yes	yes
0.079γ	yes	yes	yes
0.355γ		no	

VI. K/L-CAPTURE RATIO

The intensity of the Cs K x-ray peak in the scintillation spectrum, corrected for fluorescence yield, indicates that there are 1.7 ± 0.5 K-shell vacancies created per decay of Ba^{133}. Here it is assumed that every decay leads through the 0.079-Mev ground-state transition, and the intensity of a second gamma ray of similar energy is neglected. The number of vacancies produced by K-shell conversion of the 0.079-Mev, 0.302-Mev, and 0.355-Mev gamma rays is approximately 0.6 per decay (see Table I), and conversion of the remaining gamma rays is not expected to raise this number significantly. Hence, a considerable number of K-shell vacancies must be explained by the occurrence of K electron capture. This result is in contradiction to a previous report[3] that Ba^{133} decays mainly by L capture. The probable errors in the conversion coefficients, K/L conversion and intensity ratios combine to produce a large uncertainty in the result of a calculation of the K/L-capture ratio, but a lower limit of 1.5 can be set on this ratio.

The experimentally found preponderance of K over L capture is also expected theoretically. The observed electron capture branching ratio between the 0.434-Mev and 0.381-Mev levels indicates a decay energy of the order of 0.1 Mev to the upper level, and for this decay energy, a K/L-capture ratio of the order of 7 can be calculated.[16,17]

[16] M. E. Rose in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North Holland Publishing Company, Amsterdam, 1955), p. 273.

[17] M. E. Rose and J. L. Jackson, Phys. Rev. **76**, 1540 (1949).

VII. DISCUSSION

A tentative decay scheme for Ba^{133} is reproduced in Fig. 5, which includes the new 0.158-Mev and 0.276-Mev transitions. The sequence of these transitions is determined and their assignment confirmed by the results of Fagg,[18] who found an 0.160-Mev level in the Coulomb excitation of Cs^{133}. The coincidence results indicate that the 0.355-Mev and 0.302-Mev transitions feed the 0.079-Mev level, as indicated.

The spin and parity assignments pose a somewhat difficult problem. The ground state of Ba^{133} can be assumed to have a spin of $\frac{1}{2}$ and even parity, from the systematics of the isomerism of this isotope.[19] The Cs^{133} ground-state spin and parity presumably are $\frac{7}{2}$, even.[19,20] The 0.079-Mev ground-state transition is magnetic dipole, as indicated by the measured K-conversion coefficient, which agrees with the theoretical value[21] of 1.66 for $M1$. The 0.079-Mev level of Cs^{133} therefore presumably has a spin of $\frac{5}{2}$ and even parity. The 0.158-Mev gamma ray is not observed in the decay of Xe^{133}, which has a ground-state assignment of $\frac{3}{2}$, even, but is obtained in Coulomb excitation,[18] so that the 0.158-Mev level can have a spin of 7/2, 9/2, or 11/2, with even parity. The lower limits, within the probable errors, of the measured K-conversion coefficients of the 0.302 and 0.355-Mev gamma rays include the theoretical values[21] of 0.038 and 0.022 for magnetic dipole and electric quadrupole transitions, respectively. These assignments would make the 0.381-Mev level $\frac{3}{2}$, even and the 0.434-Mev level $\frac{1}{2}$, even, leading to allowed electron capture to both levels from the ground state of Ba^{133}. In the de-excitation of the 0.434-Mev level, there would then be competition between three gamma rays: a 0.053-Mev magnetic dipole, a 0.355-Mev electric quadrupole, and the 0.276-Mev transition which would be magnetic octopole, electric 2^4-pole, or magnetic 2^5-pole, depending upon the spin of the 0.158-Mev level.

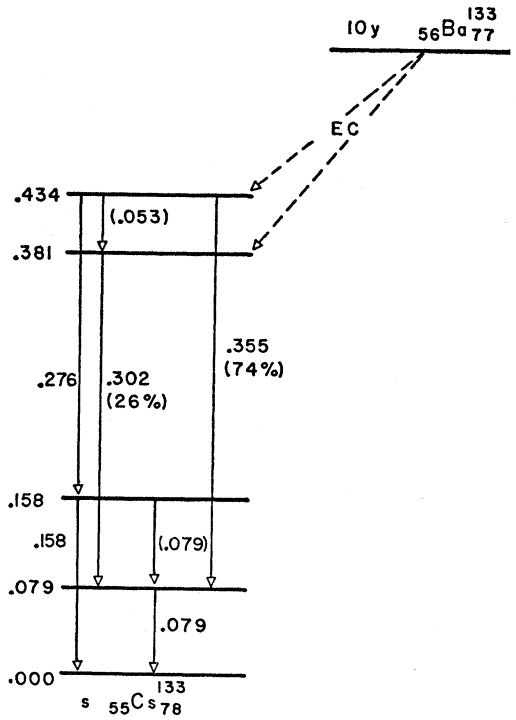

FIG. 5. Proposed decay scheme for Ba^{133}. (Energies in Mev.)

ACKNOWLEDGMENTS

The authors wish to thank Professor A. C. Helmholz, of the University of California, who kindly provided the cyclotron bombardment for the production of the Cd^{109} comparison source. Sincere thanks are due Dr. C. L. McGinnis, of the Nuclear Data Group, National Research Council, who read an early draft of this paper and supplied many valuable comments on the interpretation of the data. Helpful discussions with Dr. H. T. Easterday, of this laboratory, are gratefully acknowledged.

[18] L. W. Fagg, Bull. Am. Phys. Soc. Ser. II, 2, 207 (1957).
[19] M. Goldhaber and R. D. Hill, Revs. Modern Phys. 24, 217 (1952).
[20] P. F. A. Klinkenberg, Revs. Modern Phys. 24, 63 (1952).
[21] L. A. Sliv and I. M. Band, Report of Acad. Sci. U.S.S.R., 1956 [translation: University of Illinois Report 57 ICC K1].