

Internal-Conversion-Electron Study of the Decay of Ba<sup>133</sup> to Cs<sup>133</sup>

HANS J. HENNECKE, J. C. MANTHURUTHIL, AND O. BERGMAN

*Aerospace Research Laboratories,\* Wright-Patterson Air Force Base, Ohio*

AND

C. R. COTHERN

*University of Dayton, Dayton, Ohio*

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The 40- to 500-keV region of the internal-conversion-electron spectrum from the decay of Ba<sup>133</sup> has been investigated with an iron-core,  $\pi\sqrt{2}$  double-focusing spectrometer. Transitions of 383.79(7), 355.92(6), 302.78(5), 276.35(5), 223.15(5), 160.58(4), 80.990(22), 79.622(18), and 53.152(20) keV have been observed, and their multiplicities have been determined from measurements of the conversion-electron intensity ratios— $L_1/L_2/L_3$  and/or  $K/L$ . No new lines with intensity greater than 2.3% of the 356K line were found. The 437K line was found to have an intensity less than 0.01% of the 356K line. The multiplicities obtained for the nine observed transitions were compared with previous measurements of  $\alpha_K$  and  $\gamma$ - $\gamma$  angular correlations. This comparison resulted in unique spin-parity assignments of  $\frac{7}{2}^+$ ,  $\frac{5}{2}^+$ ,  $\frac{3}{2}^+$ ,  $\frac{3}{2}^+$ , and  $\frac{1}{2}^+$  for the ground state and the 80.997(6)-, 160.58(4)-, 383.79(7)-, and 436.93(6)-keV levels, respectively, in Cs<sup>133</sup>. The measurements of  $K/L_3$  and  $L_1/L_3$  for the 161-keV transition are consistent with previous 276-161  $\gamma$ - $\gamma$  directional-correlation measurements only if penetration effects are included with a  $\lambda$  value of  $45 \pm 10$  or  $180 \pm 20$ , where  $\lambda$  is defined as the ratio of the penetration matrix element to the normal  $\gamma$ -ray matrix element.

## INTRODUCTION

THE electron-capture decay of the long-lived isotope Ba<sup>133</sup> is known to populate, either directly or through intermediate electromagnetic transitions, four excited states in Cs<sup>133</sup>. The presently accepted level scheme (Fig. 17) is due to the work of Hayward *et al.*,<sup>1</sup> Langevin,<sup>2</sup> Crasemann *et al.*,<sup>3</sup> Gupta *et al.*,<sup>4</sup> and Koicki *et al.*<sup>5</sup> Primarily on the basis of coincidence measurements, the latter three groups<sup>3,4,5</sup> assigned energy levels at 81, 160, 380, and 437 keV. The existence of the first three of these levels has been verified in Coulomb-excitation work.<sup>6,7</sup> The ground-state spin value of  $\frac{7}{2}$  has been determined by atomic-beam methods.<sup>8</sup>

More recently the properties of the low-lying levels in Cs<sup>133</sup> have been investigated by the decay of Ba<sup>133</sup>, by the decay of Xe<sup>133</sup>, and by Coulomb excitation. The many papers covering this work can be grouped according to  $\gamma$ -ray spectroscopy,<sup>9-13</sup> conversion-electron spectroscopy,<sup>11-19</sup>  $\gamma$ - $\gamma$  angular-correlation measurements,<sup>20-24</sup>  $\gamma$ - $e_K$  and  $e_K$ - $\gamma$  angular-correlation measurements,<sup>16,25-30</sup> Coulomb excitation,<sup>31,32</sup> lifetime measure-

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<sup>14</sup> F. Brown, R. L. Graham, G. T. Ewan, and J. Uhler, *Can. J. Phys.* **39**, 779 (1961).

<sup>15</sup> K. Siegbahn, C. Nordling, S.-E. Karlsson, S. Hagstrom, A. Fahlman, and I. Andersson, *Nucl. Instr. Methods* **27**, 173 (1964).

<sup>16</sup> J. E. Thun, S. Törnkvist, K. Bonde Nielsen, H. Snellman, F. Falk, and A. Mocoroa, *Nucl. Phys.* **88**, 289 (1966).

<sup>17</sup> E. Szichman, H. E. Bosch, R. Dolinkue, and A. F. Baseggio, *Bull. Am. Phys. Soc.* **11**, 748 (1966).

<sup>18</sup> H. E. Bosch, E. Szichman, A. F. Baseggio, and R. Dolinkue, *Bull. Am. Phys. Soc.* **11**, 748 (1966).

<sup>19</sup> L. D. Hendrick and F. T. Avignone III, Program International Conference on Nuclear Physics, Gatlinburg, Tennessee, 1966 (unpublished), p. 40 (Sept. 12).

<sup>20</sup> E. Bodenstedt, H. J. Körner, and E. Matthias, *Nucl. Phys.* **11**, 584 (1959).

<sup>21</sup> F. M. Clikeman and M. G. Stewart, *Phys. Rev.* **117**, 1052 (1960).

<sup>22</sup> A. P. Arya, *Phys. Rev.* **122**, 549 (1961).

<sup>23</sup> F. Münnich, K. Fricke, and U. Wellner, *Z. Physik* **174**, 68 (1963).

<sup>24</sup> L. I. Yin and M. L. Wiedenbeck, *Nucl. Phys.* **54**, 86 (1964).

<sup>25</sup> B. N. Subba Rao, *Nucl. Phys.* **27**, 28 (1961).

<sup>26</sup> B. N. Subba Rao, *Proc. Indian Acad. Sci., Sect. A* **55**, 99 (1962).

<sup>27</sup> B. N. Subba Rao, *Nucl. Phys.* **68**, 270 (1965).

<sup>28</sup> R. H. Othaz, H. Vignau, and J. Maranon, in *Internal Conversion Processes*, edited by J. H. Hamilton (Academic Press Inc., New York, 1966), p. 450.

<sup>29</sup> M. R. Meder, H. E. Williams, and F. E. Durham, *Bull. Am. Phys. Soc.* **11**, 395 (1966).

<sup>30</sup> F. T. Avignone III, C. H. Braden, E. T. Patronis, Jr., and L. D. Wyly, *Nucl. Phys.* **80**, 314 (1966).

<sup>31</sup> D. G. Alkhazov, V. D. Vasilev, G. M. Gusinskiĭ, I. Kh. Lemberg, and V. A. Nabichvrishvili, *Izv. Akad. Nauk SSSR, Ser. Fiz.* **28**, 1683 (1964), [English transl.: *Bull., Acad. Sci. USSR, Phys. Ser.* **28**, 1575 (1964)].

<sup>32</sup> D. G. Alkhazov, K. I. Erokhina, and I. Kh. Lemberg, *Izv. Akad. Nauk SSSR, Ser. Fiz.* **28**, 1667 (1964), [English transl.: *Bull. Acad. Sci. USSR, Phys. Ser.* **28**, 1575 (1964)].

\* An element of the Office of Aerospace Research, United States Air Force.

<sup>1</sup> R. W. Hayward, D. D. Hoppes, and H. Ernst, *Phys. Rev.* **93**, 916 (1954).

<sup>2</sup> M. Langevin, *Compt. Rend* **238**, 1310 (1954); **240**, 289 (1955); *Ann. Phys. (N.Y.)* **1**, 57 (1956).

<sup>3</sup> Bernd Crasemann, J. G. Pengra, and I. E. Lindstrom, *Phys. Rev.* **108**, 1500 (1957).

<sup>4</sup> R. K. Gupta, S. Jha, M. C. Joshi, and B. K. Madan, *Nuovo Cimento* **8**, 48 (1958).

<sup>5</sup> S. D. Koicki, A. M. Mijatovic, and J. M. Simic, *Bull. Inst. Nucl. Sci. Boris Kidrich (Belgrade)* **8**, 1 (1958).

<sup>6</sup> G. M. Temmer and N. P. Heydenburg, *Phys. Rev.* **104**, 967 (1956).

<sup>7</sup> L. W. Fagg, *Phys. Rev.* **109**, 100 (1958).

<sup>8</sup> V. W. Cohen, *Phys. Rev.* **46**, 713 (1934).

<sup>9</sup> M. G. Stewart and D. C. Lu, *Phys. Rev.* **117**, 1044 (1960).

<sup>10</sup> M. K. Ramaswamy, W. L. Skeel, and P. S. Jastram, *Nucl. Phys.* **16**, 619 (1960).

<sup>11</sup> P. Erman and Z. Sujkowski, *Arkiv Fysik* **20**, 209 (1961).

<sup>12</sup> K. C. Mann and R. P. Chaturvedi, *Can. J. Phys.* **41**, 932 (1963).

<sup>13</sup> E. B. Nieschmidt, C. E. Mandeville, L. D. Ellsworth, and D. D. Bornemeier, *Phys. Rev.* **136**, B597 (1964).

ments,<sup>20,33-36</sup> and magnetic-moment measurements.<sup>20,37,38</sup> As a result of arguments based on the shell model, angular-correlation data, relatively crude conversion-coefficient measurements,  $K/(L+M)$  ratios, and  $\beta$ -decay branching, most authors agree that the probable spin sequence, in the order of increasing energy from the ground state, is:  $\frac{7}{2}$ ,  $\frac{5}{2}$ ,  $\frac{3}{2}$ ,  $\frac{1}{2}$ .

Concerning the second and fourth excited states, there exists substantial disagreement. Koicki *et al.*<sup>5</sup> favor a spin of  $\frac{3}{2}$  for the 161-keV level using arguments about gamma-ray intensities and conversion coefficients. Stewart and Lu<sup>9</sup> found an electron-capture branching of 13% to this state. Since the ground state of Ba<sup>133</sup> is, presumably, an  $s_{1/2}$  state,<sup>39,40</sup> they felt that they also could not assign a spin greater than  $\frac{3}{2}$  to the 161-keV level. On the basis of his angular-correlation measurements of the 80-81 keV gamma-gamma cascade, Arya<sup>22</sup> likewise suggests spin  $\frac{3}{2}$ . Münnich *et al.*<sup>23</sup> point out that Arya's data are also consistent with a spin of  $\frac{5}{2}$ . Further, they show that the sign of the  $A_4$  term of this correlation can, in principle, decide between the two possibilities,  $\frac{5}{2}$  and  $\frac{3}{2}$ . Unfortunately the value of the  $A_4$  term is very small while the relative errors are large. Consequently neither group can assign a unique spin value; although the data of Münnich *et al.* favors  $\frac{5}{2}$ . Bodenstedt *et al.*<sup>20</sup> and Yin and Wiedenbeck<sup>24</sup> find from their angular-correlation measurements of the 276-161 keV gamma-gamma cascade no possibility for a spin of  $\frac{3}{2}$  for the 161-keV level, and suggest instead a spin of  $\frac{5}{2}$ . Also the possibility of a spin of  $\frac{3}{2}$  is consistent with the data if the fourth excited state has a spin of  $\frac{3}{2}$  and if one allows admixture of octupole and 2<sup>+</sup>-pole radiation for the primary transitions.<sup>20</sup>

The possibility of a  $\frac{3}{2}$  spin for the 437-keV level suggested by Bodenstedt *et al.*<sup>20</sup> is supported to some extent by the findings of Subba Rao.<sup>25-27</sup> Supplementing  $\gamma$ - $\gamma$  angular-correlation data with  $\gamma$ -conversion-electron angular-correlation results, he finds that the 356-81 keV cascade is best described by the spin sequence  $\frac{3}{2}$ - $\frac{5}{2}$ - $\frac{7}{2}$ . In that case the 356-keV transition would be a mixture of 70%  $M1$ +30%  $E2$ . The value  $\frac{1}{2}$  for the spin of the 437-keV level is consistent with gamma-

gamma angular-correlation data,<sup>20-22</sup> conversion-coefficient measurements,<sup>4,16,18</sup> electron-capture branching,<sup>4,9,10</sup> and Coulomb-excitation results.<sup>7</sup>

The following facts prompted us to undertake the present investigation. All transitions have been assumed or stated to be  $M1$ ,  $E2$ , or  $M1+E2$  character, but the mixing ratios reported by different authors have not agreed very well. The few  $K$ -conversion coefficient measurements that exist are often far from decisive, since the difference between the multiplicities is not very great in this case. However, as the  $L$ -subshell ratios are quite sensitive to the multipolarity mixing, it was felt that it would be valuable to study these ratios. It should, for instance, be fairly easy to distinguish between the case of pure  $E2$  for the 356-keV transition as given by most authors, and the case of 70%  $M1$ +30%  $E2$  as given by Subba Rao.<sup>25</sup> Also, by measuring the conversion intensities accurately the question about the electron-capture intensity to the 161-keV state could be resolved.

Ba<sup>133</sup>, having a long half-life of 7-11 years<sup>41,42</sup> and several conversion lines in the range 50-350 keV, could constitute a fairly good calibration source for  $\beta$ -ray spectrometers. We therefore decided to undertake in addition a careful energy calibration of the conversion lines against the accurately known standard lines in Au<sup>199</sup>, I<sup>131</sup>, and Cs<sup>137</sup>.

## APPARATUS AND TECHNIQUE

### Source Preparation

The primary source activity was obtained commercially<sup>43</sup> as a solution of BaCl<sub>2</sub> in 1N HCl containing 1 mCi of Ba<sup>133</sup>. Since the specific activity of the stock item obtained was relatively poor (about 1 mCi/mg), sources of different thicknesses were prepared for high- and low-energy studies. The sources were produced by the technique of molecular plating.<sup>44</sup> After removing the HCl by evaporating the stock solution to dryness, the residue was dissolved in 1-2 drops of water and added to the 15 ml of dry isopropanol in the Teflon molecular plating cell. The source areas, 2×20 mm<sup>2</sup> and 4×20 mm<sup>2</sup>, were defined by covering the aluminum source backing foils (4 mg/cm<sup>2</sup>) with appropriate Teflon masks. Using a plating voltage of 420 V, 75% of the activity was typically deposited in about one hour. The sources appeared uniform and the deposit, a white layer for the thickest source, adhered well to the aluminum backing.

<sup>33</sup> W. Flauger and H. Schneider, *Atomkernenergie* **8**, 453 (1963).

<sup>34</sup> G. A. Vartapetyan, A. G. Khudaverdyan, and T. A. Garibyan, *Zh. Eksperim. i Teor. Fiz.* **45**, 1720 (1964) [English transl.: *Soviet Phys.—JETP* **18**, 1178 (1964)].

<sup>35</sup> G. A. Vartapetyan, T. A. Garibyan, N. A. Demekhina, and E. G. Muradyan, *Izv. Akad. Nauk, Ser. Fiz.* **28**, 1657 (1964) [English transl.: *Bull. Acad. Sci. USSR, Phys. Ser.* **28**, 1550 (1964)].

<sup>36</sup> T. Scharbert, *At. Kozlemen* **6**, 137 (1964).

<sup>37</sup> Y. K. Agarwal, C. V. K. Baba, and S. K. Bhattacharjee, *Nucl. Phys.* **58**, 651 (1964).

<sup>38</sup> Y. K. Agarwal, C. V. K. Baba, and S. K. Bhattacharjee, *Nucl. Phys.* **63**, 685 (1965).

<sup>39</sup> R. D. Hill, G. Scharff-Goldhaber, and M. McKeown, *Phys. Rev.* **84**, 382 (1951).

<sup>40</sup> J. E. Thun, S. Törnkvist, F. Falk, and H. Snellman, *Nucl. Phys.* **67**, 625 (1965).

<sup>41</sup> *Nuclear Data Sheets*, compiled by K. Way *et al.*, (Printing and Publishing Office, National Academy of Science, National Research Council, Washington, D. C., 25), NRC 61-2-91.

<sup>42</sup> E. I. Wyatt, S. A. Reynolds, T. H. Handley, W. S. Lyon, and H. A. Larker, *Nucl. Sci. Eng.* **11**, 74 (1961).

<sup>43</sup> Oak Ridge National Laboratories, Oak Ridge, Tennessee.

<sup>44</sup> W. Parker, thesis, Chalmers Technical High School, Göteborg, Sweden, 1965 (unpublished).

### Beta-Ray Spectrometer

An iron-core, double-focusing  $\beta$ -ray spectrometer of 50-cm mean radius was used in the present investigation. This instrument is similar to the spectrometer described by Pettersson *et al.*<sup>45</sup> Two modifications of importance to the present investigation were a magnetic field correction coil and a Si(Li) detector.

The focusing properties of the spectrometer have been improved by the addition of a correction coil<sup>46</sup> wound axially along the upper part of the inner vacuum tank and carrying 1.6% of the ampere-turns in the main coil. This coil has the effect of strengthening the upper part of the field relative to the lower part by a few tenths of a percent. With an aperture solid angle  $\Omega/4\pi=0.25\%$ , 2-mm-wide source, and 2-mm-wide detector slit, the momentum resolution obtained is 0.14%. This value is very close to what one can expect theoretically with this type of spectrometer.

The electrons were detected using a commercially obtained Si(Li) detector<sup>47</sup> with a rectangular cross section of  $5 \times 25$  mm<sup>2</sup> and 2-mm depth. The detector was cooled by means of a liquid-nitrogen cryostat. During the course of the investigation two separate electronic systems, one a standard vacuum tube system<sup>48</sup> and the second an all solid-state system<sup>49</sup> were utilized. The typical energy resolution of the detection system was 10–30 keV full width at half-maximum (FWHM).

The use of an integral discriminator to eliminate the effects of detector and electronics noise coupled with electron backscatter effects resulted in a strongly energy-dependent efficiency for the counting of low-energy electrons (less than, say, 100 keV). The efficiency of the detection system was determined by examining pulse-height spectra resulting from the detection of monoenergetic electrons selected by the spectrometer. The efficiency of the detection system was also checked by the known intensities of the conversion lines<sup>50</sup> from Au<sup>199</sup> and by measurements of the shape of the Co<sup>60</sup>  $\beta$  continuum.<sup>51</sup>

Conversion line intensities were obtained by measuring the line areas with a planimeter, dividing each area by the appropriate  $B\rho$  value for the line, and correcting for the detector efficiency. The intensities of most of the lines were measured several times in independent

experiments utilizing different sources, different spectrometer resolution settings, and even different detector electronics in some cases. As a result of all these measurements and the studies of detector efficiency, we feel that the systematic error for conversion line relative intensities is less than 10%.

### Energy Measurements

The energies of all  $K$ -conversion lines (except the 53 $K$ , 223 $K$ , and 437 $K$  lines) and the 53 $L_1$  line were carefully calibrated against the well-known lines from Ba<sup>133</sup> (81 $K$ ), Au<sup>199</sup> (50 $L_1$ , 158 $K$ , 208 $K$ , 208 $L_1$ ), I<sup>131</sup> (284 $K$ , 364 $K$ ), and Cs<sup>137</sup> (661 $K$ ). The appropriate calibration energies and momenta were obtained from Siegbahn *et al.*<sup>15,52</sup> The binding energies for Cs<sup>133</sup> obtained from Hagström<sup>53</sup> are  $B(K)=35.989$ ,  $B(L_1)=5.717$ ,  $B(L_2)=5.363$  and  $B(L_3)=5.016$  keV. Since most of the  $K$ -conversion lines in the decay of Ba<sup>133</sup> have energies very close to those of the calibration lines used, very small systematic errors for the energy measurements are expected. Care was taken to compensate for the small radial asymmetry of the sources. For a given source this effect was determined by observing the shift of a prominent line when the source was inverted from its normal orientation. The crossing point of the extrapolated line edges was used for momentum definition.<sup>54</sup>

Three separate calibration runs were performed with 2-mm-wide sources, 2-mm-wide detector slit opening, and various baffle settings. In each case the instrumental momentum resolution, as determined from the higher energy lines, was 0.14%. Broadening of the lower-energy lines from source thickness and backscattering effects was observed. For example, for the strong 2-mm Ba<sup>133</sup> source the observed linewidths of the 81 $K$  and 81 $L$  lines were 0.56% and 0.32%, respectively; while for a thinner 2-mm Ba<sup>133</sup> source the equivalent linewidths were 0.22% and 0.20%, respectively.

### $L$ -Subshell Analysis

The percentage difference in momentum between the  $L_1$  and  $L_3$  conversion lines for transitions in Cs<sup>133</sup> are indicated in Fig. 1. For the transitions with energy greater than or equal to 161 keV, the  $L_1$ - $L_3$  separations are close to the practical limits of resolution as determined by the relative transition intensity, the spectrometer transmission, and the thickness and specific activity of the available sources. Figure 1 also indicates the experimental resolutions obtained for each  $L$ -subshell measurement. It is clear from this information

<sup>45</sup> H. Pettersson, Y. Grunditz, G. Bäckström, O. Bergman, S. Antman, and E. Aasa, *Arkiv Fysik*, **29**, 61 (1965).

<sup>46</sup> H. J. Hennecke, O. Bergman, and J. C. Manthuruthil, *Bull. Am. Phys. Soc.* **11**, 605 (1966).

<sup>47</sup> Model LR-125-2.0-90, made by Simtec Ltd., Montreal, Quebec.

<sup>48</sup> Models N358A preamplifier, N371 linear amplifier and integral discriminator, and N-375 biased amplifier system made by Hamner Electronics Co.

<sup>49</sup> Models 109 preamplifier, 410 linear amplifier, 210 detector bias supply made by ORTEC, and a N685 pulse-height analyzer made by Hamner.

<sup>50</sup> G. Bäckström, O. Bergman, and J. Burde, *Nucl. Phys.* **7**, 263 (1958).

<sup>51</sup> F. Bonhoeffer, *Z. Physik* **154**, 62 (1959).

<sup>52</sup> K. Siegbahn, in *Alpha-, Beta-, and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland Publishing Company, Amsterdam, 1965), p. 198.

<sup>53</sup> S. Hagström, C. Nordling, and K. Siegbahn, in *Alpha-, Beta-, and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland Publishing Company, Amsterdam, 1965), p. 845.

<sup>54</sup> C. DeVries, *Nucl. Phys.* **18**, 428 (1960).

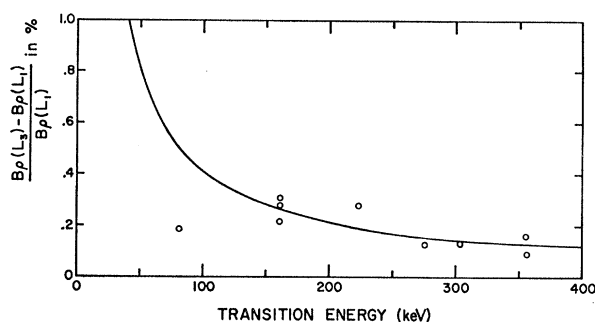


FIG. 1. The  $L_1$ - $L_3$  momentum separation in percent as a function of transition energy. The circles indicate the experimental resolution (including line broadening from source effects) obtained in the  $L$ -subshell measurements.

that, in general, the resolution utilized was insufficient to resolve completely the individual lines but good enough to indicate broadening or other structure of the composite line.

For purposes of analysis a computer code was prepared which would construct a composite line utilizing theoretical  $L$ -subshell conversion coefficients,<sup>55</sup> the  $(L+1)$ -pole/ $L$ -pole amplitude mixing ratio ( $\delta^2$ ), the  $L_1$ - $L_2$ - $L_3$  separations determined from the atomic binding energies,<sup>53</sup> and a reference line-shape obtained from nearby  $K$  lines. The normalization factor for this calculated composite line together with two constants defining a linear background are then determined by least-squares techniques. The resulting goodness of fit is checked by examining the value of  $Q^2$  defined as:

$$Q^2 = \frac{1}{n-5} \sum_{i=1}^n \frac{[W(B_i) - W^*(B_i)]^2}{W(B_i)}, \quad (1)$$

where  $W(B_i)$  is the experimental counting rate at a given spectrometer field setting  $B_i$ , and  $W^*(B_i)$  is the corresponding value for the composite line and background computed as mentioned above. Here it is assumed that  $W(B_i)$  follows Poisson statistics and that the reference line shape is known exactly. Of the five degrees of freedom subtracted from the total number of data points  $n$ , three are for the normalization constants mentioned above, one is for the mixing ratio  $|\delta|$  and one is for the magnetic field setting  $B_1$  of the  $L_1$  line. The minimum value of  $Q^2$  is referred to as  $\chi^2$ .

In practice  $Q^2(|\delta|)$  is calculated at values of the parameter  $\arctan |\delta|$  equally spaced between  $0^\circ$  and  $90^\circ$ . For a given mixing ratio  $|\delta|$  the value of the magnetic field setting  $B_1$  of the  $L_1$  line is varied within the range  $B_1^0 \pm \Delta B_1$  until a minimum value of  $Q^2$  is found. The quantity  $B_1^0$  is the magnetic-field setting for the  $L_1$  line as determined from the field setting for the  $K$  line. The quantity  $\Delta B_1$  represents the error of

$B_1^0$ . The confidence limits for the mixing ratio  $|\delta_1| \leq |\delta| \leq |\delta_2|$  arising from statistical considerations only are determined by the condition:

$$Q^2(|\delta_i|) = \chi^2 + 1/(n-5). \quad (2)$$

Here  $i$  has the values 1 or 2 corresponding to the two solutions near the minimum  $\chi^2$  of the quadratic function  $Q^2(|\delta|)$ . It has been shown<sup>56</sup> that the error obtained in this way properly accounts for any correlation between two variables such as  $|\delta|$  and  $B_1$ .

## RESULTS

### Internal-Conversion Spectrum

Internal-conversion lines were observed from all but one (437-keV) of the ten possible transitions between the five known states of  $\text{Cs}^{133}$  populated by the decay of  $\text{Ba}^{133}$ . The electron spectrum from 40–500 keV was examined in order to determine the upper intensity limits for possible new weak lines as well as for the  $K$  line corresponding to the transition from the 437-keV level to the ground state. No new lines were found nor was the 437K line observed. The intensity limits are summarized in Table I. These limits, of course, do not hold near the known conversion lines where somewhat stronger lines could be obscured. If the 437-keV transition is indeed  $M3$  as generally accepted, the present intensity limit  $I_{437K}/I_{356K} \leq 1 \times 10^{-4}$  indicates that the 437-keV  $\gamma$ -ray intensity is 500 times weaker than the best previous estimate.<sup>4</sup> If this transition is  $E2$ , than the  $\gamma$ -ray intensity is 50 times weaker than previously found. Figure 2 is a composite electron spectrum covering the energy region 40–500 keV and made up of separate spectra taken during the intensity-limit investigations. The individual runs have been suitably normalized, where appropriate, in order to account for the different transmission settings used. The spectra shown have not been corrected for the detector efficiency variation with energy.

TABLE I. Intensity limits for weak conversion lines from the decay of  $\text{Ba}^{133}$ .

Electron energy range (keV)	Intensity limit <sup>a</sup> (%)
54–71	2.3
88–99	0.70
99–118	0.55
117–158	0.15
150–230	0.07
210–319	0.09
319–347	0.05
355–493	0.03
437 K-line	0.01

<sup>a</sup> The intensity limit is defined as  $300N_{\delta}^{1/2}/N_{356K}$ , where  $N_{\delta}$  is the background count rate and  $N_{356K}$  is the peak count rate for the 356K line.

<sup>55</sup> I. M. Band, M. A. Listengarten, and L. A. Sliv, in *Alpha-, Beta-, and Gamma-Ray Spectroscopy*, edited by K. Siegbahn, (North-Holland Publishing Company, Amsterdam, 1965), Appendix G.

<sup>56</sup> D. D. Watson, thesis, University of Kansas, 1965 (unpublished).

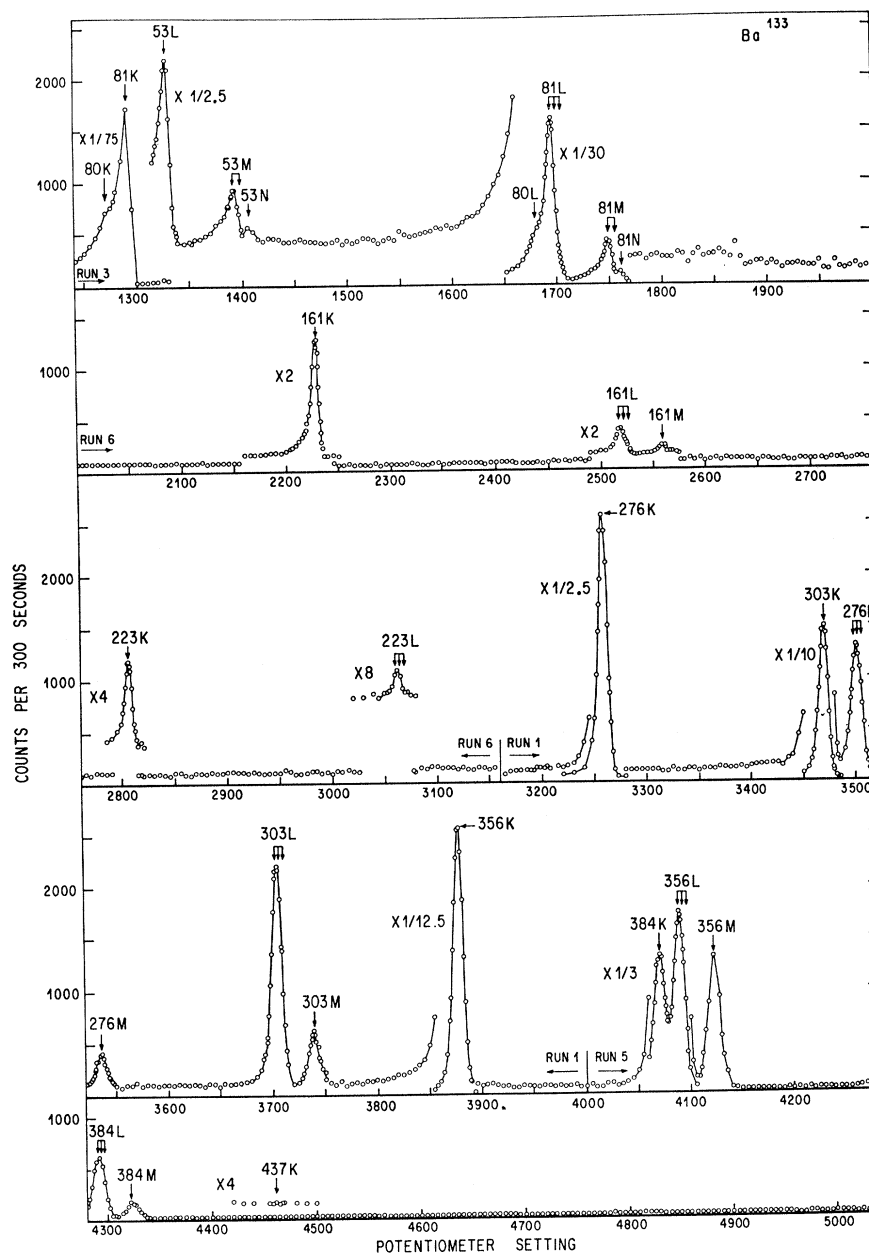


FIG. 2. Composite internal-conversion-electron spectrum showing the region 40–500 keV in the decay of  $\text{Ba}^{133}$ . This spectrum is composed of four separate runs which have been suitably normalized, where appropriate, to account for the different transmission settings used.

### Energy and Intensity Measurements

The energies of the 81K and 81L lines were calibrated relative to the 50M and 158K lines of  $\text{Au}^{199}$ . The resulting transition energy  $E_\gamma = 80.990 \pm 0.022$  keV is in good agreement with the more accurate measurement  $E_\gamma = 80.997 \pm 0.006$  keV of Siegbahn *et al.*<sup>15</sup> The 53L<sub>1</sub> and 80K lines were calibrated relative to the 81K line.<sup>15</sup> The energies of the 161K, 276K, 303K, 356K, and 384K lines were measured in two independent calibration experiments. The energy of the 223K line was not measured during these calibration experiments. However, in a later experiment for the 161L-subshell

investigation, the position of the 223K line was obtained relative to the 161K, 161L, and 356K lines. The results of these direct energy measurements are listed in Table II.

The energy of the unobserved 437-keV transition can be obtained from the cascades  $E_\gamma(356) + E_\gamma(81)$  and  $E_\gamma(384) + E_\gamma(53)$ . Using the transition energies from Table II we obtain  $E_\gamma(437) = 436.93 \pm 0.06$  keV. In a similar way the energy of the 223-keV transition can also be obtained from  $E_\gamma(303) - E_\gamma(80)$  and  $E_\gamma(276) - E_\gamma(53)$ . Combining these values with the direct measurement we get  $E_\gamma(223) = 223.15 \pm 0.05$  keV.

TABLE II. Energy and intensity measurements.

Assignment	Electron energy keV	Transition energy keV	Relative conversion intensities	$K/L$	$L/(M+N)$
80K <sup>a</sup>	43.633±0.018	79.622±0.018	26 ± 4	6.4 ±1.2	...
81K	45.016±0.023	81.005±0.023	292 ±36	5.72±0.73	3.6±0.4
53L <sub>1</sub> <sup>a</sup>	47.435±0.020	53.152±0.020	8.5 ± 1.2	...	3.1±0.3
81L <sub>1</sub>	75.258±0.021	80.975±0.021	...	...	...
161K	124.55 ±0.07	160.54 ±0.07	1.10± 0.14	4.80±0.54	4.5±0.5
223K <sup>b</sup>	187.11 ±0.08	223.10 ±0.08	0.26± 0.03	7.82±0.81	...
276K	240.36 ±0.05	276.35 ±0.05	2.51± 0.27	5.63±0.37	3.8±0.4
303K	266.79 ±0.05	302.78 ±0.05	5.29± 0.53	6.95±0.56	4.0±0.4
356K	319.93 ±0.06	355.92 ±0.06	10	6.06±0.34	3.9±0.4
384K	347.80 ±0.07	383.79 ±0.07	1.18± 0.13	6.05±0.43	4.3±0.4

<sup>a</sup> The energies of the 80K and 53L<sub>1</sub> lines were calibrated against 81K line utilizing  $E_\gamma(81) = 80.997(6)$ . (Ref. 15).

<sup>b</sup> The energy of the 223K line was measured relative to the 161L<sub>1</sub> line.

Finally the value of the energy of the 161-keV level can be improved by considering the sums  $E_\gamma(81) + E_\gamma(80)$  and  $E_\gamma(437) - E_\gamma(276)$ . In this way we get  $E_\gamma(161) = 160.58 \pm 0.04$  keV. The resulting adopted values for the transition energies are shown in Fig. 17.

The relative intensities of the  $K$ -conversion lines (except 53K and 437K) and the 53L<sub>1</sub> and 81L<sub>1</sub> lines were measured at least three times. The average intensities are summarized in Table II. The intensity errors quoted include the external error calculated from the spread of the individual measurements plus a 10% contribution for the error in the efficiency of the detection system as discussed in a previous section.

After the completion of our energy and intensity measurements and analysis, it came to our attention that Thun *et al.*<sup>16</sup> had measured the energies and intensities of the lines 53K, L<sub>1</sub>, L<sub>2</sub>, L<sub>3</sub>, 80K, 81K, 80L<sub>1</sub>, L<sub>2</sub>, L<sub>3</sub>, 161K, and 356K. They utilized an iron-free double-focusing spectrometer and normalized the energies to the 81K electron energy measured by Siegbahn *et al.*<sup>15</sup> In addition they measured the relative intensities of the 276K, 303K, 356K, and 384K lines with a lens spectrometer. For comparison, their energy and intensity values are listed together with the corresponding values obtained in the present work in Table

III. We have renormalized their intensity measurements to the intensity of the 356K line. The two sets of data are in quite good agreement.

The only other accurate energy measurements for transitions in Cs<sup>133</sup> available at this time are the values:  $E_\gamma(80) = 79.59 \pm 0.03$  keV and  $E_\gamma(81) = 80.97 \pm 0.03$  keV by Erman and Sujkowski,<sup>11</sup> and  $E_\gamma(81) = 80.99 \pm 0.02$  keV by Brown *et al.*<sup>14</sup>

#### Transition Multipolarities and Mixing Ratios

The data for the  $K/L$  and  $L$ -subshell ratios for each transition investigated were analyzed in terms of the multipolarity mixings:  $E2/M1$ ,  $M3/E2$ ,  $M2/E1$ , and  $E3/M2$ . The necessary conversion coefficients were obtained from the Sliv-Band tabulations<sup>55</sup> by linear interpolation of a log-log plot. A computer program was utilized to convert the tabulated coefficients and energies to logarithmic values and to perform the interpolation. The tabulated conversion coefficients were also plotted by hand to check the validity of our interpolation techniques. Figures 3-8 show the data for the various  $L$ -lines together with the best computer fit to the data. Curves of  $Q^2(|\delta|)$  versus  $\arctan |\delta|$  for the above listed multipolarity mixings are given

TABLE III. Energy and intensity comparison.

Transition	Transition energy (keV)		Electron intensity	
	Present work	Thun <i>et al.</i> <sup>a</sup>	Present work	Thun <i>et al.</i> <sup>a</sup>
53L <sub>1</sub>	53.152±0.020	53.17 ±0.04	8.5 ± 1.2	10.4 ± 0.5
80K	79.622±0.018	79.60 ±0.05	26 ± 4	29.3 ± 2.1
81K	80.990±0.022 <sup>b</sup>	80.997±0.006 <sup>f</sup>	292 ±36	340 ±12
161K	160.58 ±0.04 <sup>c</sup>	160.66 ±0.06	1.10± 0.14	1.70± 0.48
223K	223.15 ±0.05 <sup>d</sup>	(223.43 ±0.26)	0.26± 0.03	
276K	276.35 ±0.05	(276.43 ±0.26)	2.51± 0.27	2.65 <sup>g</sup>
303K	302.78 ±0.05	(303.09 ±0.21)	5.29± 0.53	5.10 <sup>g</sup>
356K	355.92 ±0.06	356.26 ±0.15	10	10
383K	383.79 ±0.07	(384.09 ±0.20)	1.18± 0.13	1.15 <sup>g</sup>
437K	436.93 ±0.06 <sup>e</sup>	(437.26 ±0.16)	≤0.001	

<sup>a</sup> See Ref. 16. The values in parentheses were not measured directly.

<sup>b</sup> Weighted average resulting from 81K and 81L<sub>1</sub> measurements.

<sup>c</sup> Average of values obtained from 161K measurement,  $E_\gamma(81) + E_\gamma(80)$ , and  $E_\gamma(437) - E_\gamma(276)$ .

<sup>d</sup> Average of values obtained from 223K measurement,  $E_\gamma(303) - E_\gamma(80)$ , and  $E_\gamma(276) - E_\gamma(53)$ .

<sup>e</sup> Average of  $E_\gamma(356) + E_\gamma(81)$  and  $E_\gamma(384) + E_\gamma(53)$ .

<sup>f</sup> This value for the energy of the 81-keV transition was taken from Ref. 15 and utilized by Thun *et al.* to normalize their data.

<sup>g</sup> No error limits given for the intensities of the 276K, 303K, and 384K lines in Ref. 16.

in Figs. 9-14. The resulting multipolarity mixing possibilities are listed in Table IV.

The 161L line was investigated in three separate experiments at different resolutions. The results of each of these measurements are listed in Table IV. However, only the data and analysis for the best resolution obtained for the 161L line are shown in Figs. 3 and 9, respectively. Similarly the 356L line was examined twice and only the better resolution data and analysis are presented in Figs. 8 and 14, respectively.

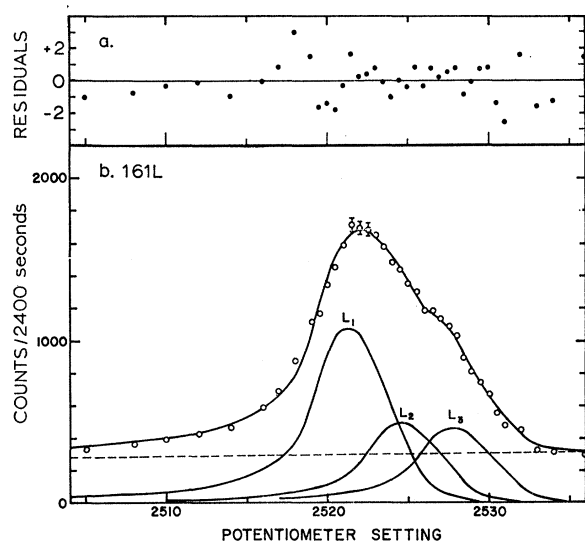


FIG. 3. 161L line with  $\Delta B\rho/B\rho=0.23\%$ ,  $\chi^2=1.46$ ,  $|\delta|=1.03$  (E2/M1). (a) The residuals,  $[W(B_i)-W^*(B_i)]/\sqrt{W(B_i)}$ , are plotted versus potentiometer setting. (b) The three L-subshell lines (solid curves), the linear background (dashed line), and the composite L-line (solid curve through the data points), all obtained by a least-square fit are shown. The error bars on the data points represent statistical errors only.

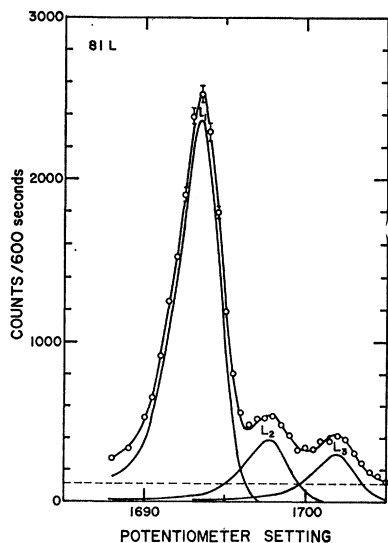


FIG. 4. 81L line with  $\Delta B\rho/B\rho=0.22\%$ ,  $\chi^2=0.54$ ,  $|\delta|=0.173$  (E2/M1). See caption, Fig. 3.

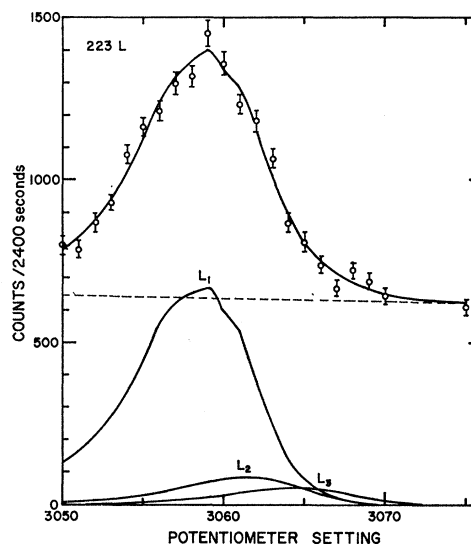


FIG. 5. 223L line with  $\Delta B\rho/B\rho=0.28\%$ ,  $\chi^2=1.51$ ,  $|\delta|=0.40$  (E2/M1). See caption, Fig. 3.

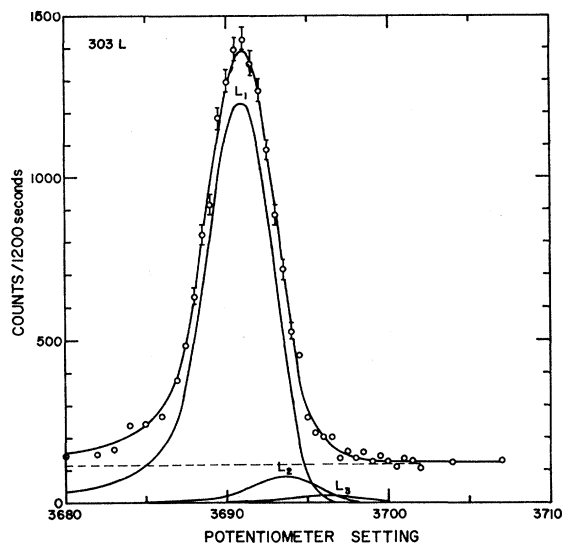


FIG. 6. 303L line with  $\Delta B\rho/B\rho=0.13\%$ ,  $\chi^2=1.55$ ,  $|\delta|=0.11$  (E2/M1). See caption Fig. 3.

It should be pointed out that the mixing ratios listed in Table IV for the 53- and 80-keV transitions were not obtained by computer fits but only by crude estimates of the  $L_1/L_2$  ratios from the data; namely,  $L_1/L_2=7.4\pm 1.3$  and  $\geq 8$ , respectively. Also the line shape used to analyze the L-subshell data for the 81-keV transition was obtained by successive trials because no nearby K line was available in the spectrum. As can be seen from Fig. 4 the resolution was nearly good enough to resolve the strong  $L_1$  line from the weak  $L_2$  component. As a first approximation the  $L_1$ -line shape, with the high-energy edge suitably extrapolated to zero, was used as a trial shape to subtract out the effects of the  $L_2$  and  $L_3$  components. This process was

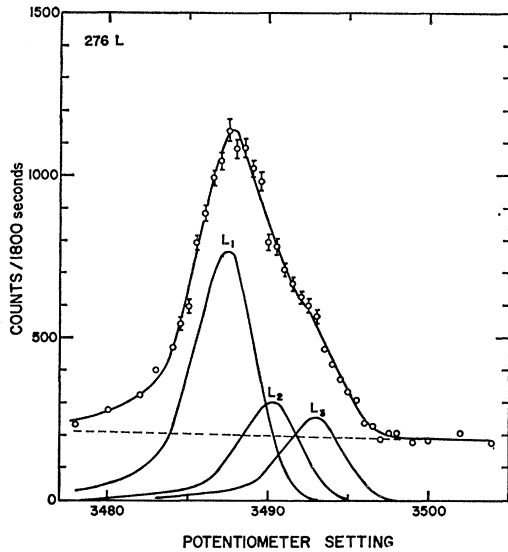


FIG. 7. 276L line with  $\Delta B\rho/B\rho=0.13\%$ ,  $\chi^2=1.17$ ,  $|\delta|=0.028$  ( $M3/E2$ ). See caption Fig. 3.

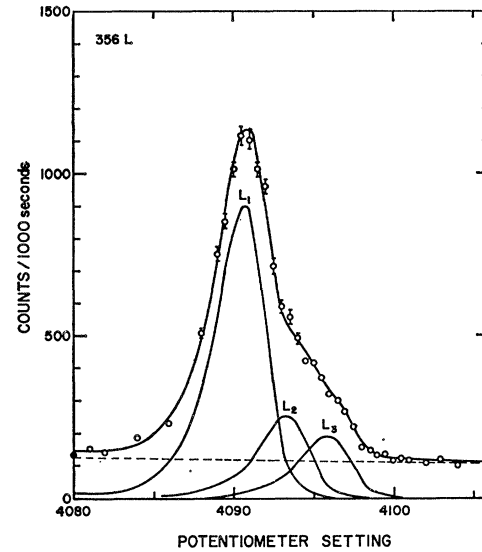


FIG. 8. 356L line with  $\Delta B\rho/B\rho=0.09\%$ ,  $\chi^2=0.90$ ,  $|\delta|=0.033$  ( $M3/E2$ ). See caption Fig. 3.

repeated until no improvement in the degree of fit was obtained.

The measured  $K/L$  ratios and the resulting values of  $|\delta|$  are also given in Table IV. The  $K/L$  ratios for the 223- and 303-keV transitions allow the possibility of  $E0$  admixture in these transitions. This fact has previously been pointed out by Nieschmidt *et al.*<sup>13</sup> for the 223-keV transition. If these transitions do indeed have  $E0$  admixtures, then the 384-, 161-, and 81-keV levels must all have the same spin, contrary to the generally accepted assignment of  $\frac{3}{2}$ ,  $\frac{5}{2}$ , and  $\frac{5}{2}$ , respectively. The possibility of  $E0+M1+E2$  mixing for

these transitions was checked with the  $L$ -subshell measurements. The conversion rates,  $L_1$ ,  $L_2$ ,  $L_3$  can be written as:

$$L_1 = N_\gamma(M1)\{\beta_{L_1}(M1) + \delta^2[\alpha_{L_1}(E2) + \epsilon^2\alpha_{L_1}(E0)]\}, \quad (3a)$$

$$L_2 = N_\gamma(M1)\{\beta_{L_2}(M1) + \delta^2[\alpha_{L_2}(E2) + \epsilon^2\alpha_{L_2}(E0)]\}, \quad (3b)$$

$$L_3 = N_\gamma(M1)\{\beta_{L_3}(M1) + \delta^2\alpha_{L_3}(E2)\}. \quad (3c)$$

Here  $\epsilon^2$  is the ratio of the rate  $N_K(E0)$  of  $K$ -shell  $E0$  conversion to the rate of  $E2$  gamma-ray emission. Also,  $\delta^2$  is the ratio of the  $E2$   $\gamma$ -ray emission rate to the  $M1$   $\gamma$ -ray emission rate  $N_\gamma(M1)$ . The  $\alpha_i(E2)$  and  $\beta_i(M1)$

TABLE IV. Multipolarity mixing from  $L$ -subshell and  $K/L$  ratios.

Transition	Measurement	Resolution <sup>c</sup> %	Multipolarity mixing: $ \delta $			
			$E2/M1$	$M3/E2$	$M2/E1$	$E3/M2$
53	$L_1/L_2$ <sup>a</sup>	0.28	$0.10 \pm 0.02$	$\geq 0.55$	$0.2_{-0.1}^{+0.4}$	$\leq 0.063$
80	$L_1/L_2$ <sup>b</sup>	0.23	$\leq 0.15$	...	...	...
	$K/L$		$\leq 0.29$	...	$\leq 0.53$	...
81	$L_1/L_2/L_3$	0.22	$0.173 \pm 0.004$	...	...	...
	$K/L$		$0.24_{-0.07}^{+0.08}$	...	$0.12_{-0.05}^{+0.09}$	...
161	$L_1/L_2/L_3$	0.32	$1.12_{-0.18}^{+0.32}$	$0.10 \pm 0.07$	...	$0.49 \pm 0.06$
	$L_1/L_2/L_3$	0.28	$1.23_{-0.15}^{+0.19}$	$0.13 \pm 0.03$	...	$0.51 \pm 0.04$
	$L_1/L_2/L_3$	0.23	$1.03_{-0.05}^{+0.06}$	$0.19 \pm 0.02$	...	$0.46 \pm 0.02$
223	$K/L$	...	$1.05 \pm 0.70$	...	...	$0.23_{-0.14}^{+0.10}$
	$L_1/L_2/L_3$	0.28	$0.40_{-0.18}^{+0.14}$	...	$\leq \infty$	$0.11 \pm 0.11$
276	$K/L$	...	$\leq 0.38$	...	$\leq 0.16$	...
	$L_1/L_2/L_3$	0.13	$6.8_{-3.2}^{+0.0}$	$0.03_{-0.03}^{+0.05}$	...	$0.82_{-0.05}^{+0.04}$
303	$K/L$	...	$2.6_{-1.2}^{+0.0}$	$\leq 0.12$	...	$0.36_{-0.14}^{+0.13}$
	$L_1/L_2/L_3$	0.13	$0.11_{-0.11}^{+0.07}$	...	$\geq 1.4$	$\leq 0.03$
356	$K/L$	...	$0.62_{-0.46}^{+0.52}$	...	$0.26_{-0.15}^{+0.96}$	...
	$L_1/L_2/L_3$	0.16	$\geq 3.9$	$\leq 0.11$	...	$1.01 \pm 0.11$
384	$L_1/L_2/L_3$	0.09	$6.2_{-2.4}^{+0.0}$	$0.033_{-0.033}^{+0.061}$	...	$0.86_{-0.04}^{+0.03}$
	$K/L$	...	$10_{-8.2}^{+0.0}$	$\leq 0.21$	...	$0.40_{-0.19}^{+0.17}$
	$K/L$	...	$\geq 1.98$	$0.13_{-0.13}^{+0.32}$	...	$0.48_{-0.25}^{+0.23}$

<sup>a</sup>  $L_1/L_2=7.4 \pm 1.3$ .

<sup>b</sup>  $L_1/L_2 \geq 8$ .

<sup>c</sup> The resolution values refer to the observed momentum resolution including source thickness, backscatter, and instrumental effects.



are the usual tabulated conversion coefficients. The quantities  $\alpha_{L_1}(E0)$  and  $\alpha_{L_2}(E0)$  are defined to be:

$$\alpha_{L_1}(E0) = \frac{L_1}{L_2}(E0)\alpha_{L_2}(E0), \quad (4a)$$

$$\alpha_{L_2}(E0) = \left\{ \frac{K}{L}(E0) \left[ 1 + \frac{L_1}{L_2}(E0) \right] \right\}^{-1}. \quad (4b)$$

The values of  $K/L$  and  $L_1/L_2$  for  $E0$  transitions can be obtained from the graphs given by Church and Wenner.<sup>57</sup>

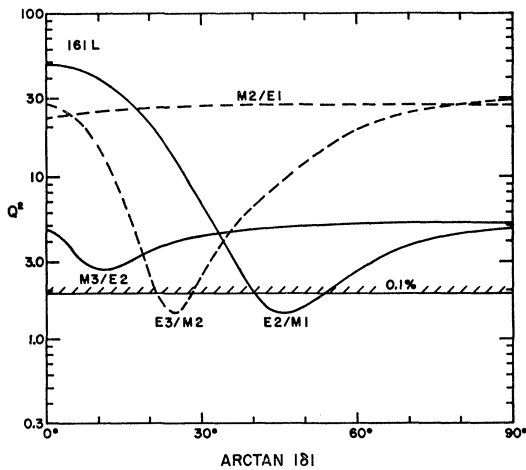


FIG. 9.  $Q^2$  versus  $\arctan |\delta|$  for the 161L line with  $\Delta B\rho/B\rho = 0.23\%$ . The 0.1% confidence limit is also shown.

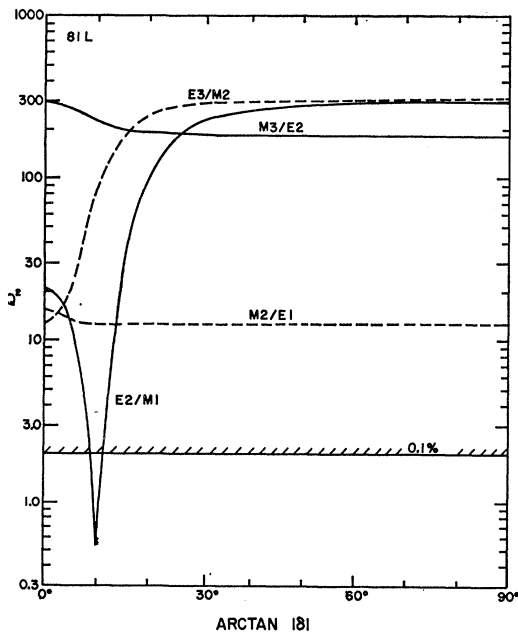


FIG. 10.  $Q^2$  versus  $\arctan |\delta|$  for the 81L line with  $\Delta B\rho/B\rho = 0.22\%$ .

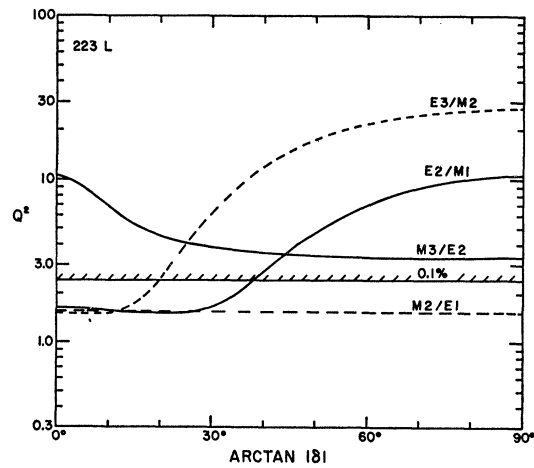


FIG. 11.  $Q^2$  versus  $\arctan |\delta|$  for the 223L line with  $\Delta B\rho/B\rho = 0.28\%$ .

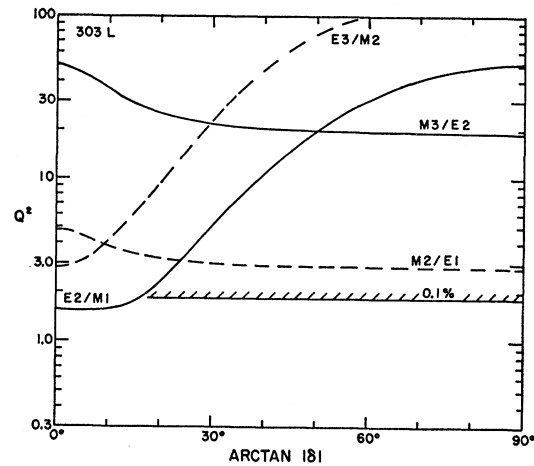


FIG. 12.  $Q^2$  versus  $\arctan |\delta|$  for the 303L line with  $\Delta B\rho/B\rho = 0.13\%$ .

While the  $L$ -subshell data for these transitions are more restrictive for the allowable values of  $\epsilon^2$  and  $\delta^2$  than the  $K/L$  data, they are by no means decisive. For the 223-keV transition, the  $L$ -subshell measurement restricts  $\epsilon^2 \leq 0.7$  and  $\delta^2 \geq 0.2$ . The best fit to the  $L$ -subshell data corresponds to  $\epsilon^2 = 0$  and  $|\delta| = 0.40 \pm 0.14$ . For  $\epsilon^2 = 0$ , the  $K/L$  measurement gives  $|\delta| \leq 0.4$ . We cannot by these measurements alone rule out the possibility that there exists some  $E0$  admixture with a corresponding increase in the  $E2$  intensity.

In the case of the 303-keV transition  $K/L$  measurement is not at all restrictive; the whole  $\epsilon^2 - \delta^2$  space is spanned by an error limit less than two standard deviations in the value of  $K/L$ . The  $L$ -subshells allow a solution for  $\epsilon^2 = 0$  and  $|\delta| = 0.11_{-0.11}^{+0.07}$ ; however, a solution with somewhat lower  $\chi^2$  exists in the region  $0.22 \leq \epsilon^2 \leq 0.49$  and  $|\delta| \geq 0.36$ . Hence the present measurements do not, in themselves, rule out  $E0$  admixture for the 303-keV transition.

<sup>57</sup> E. L. Church and J. Wenner, Phys. Rev. **103**, 1035 (1956).

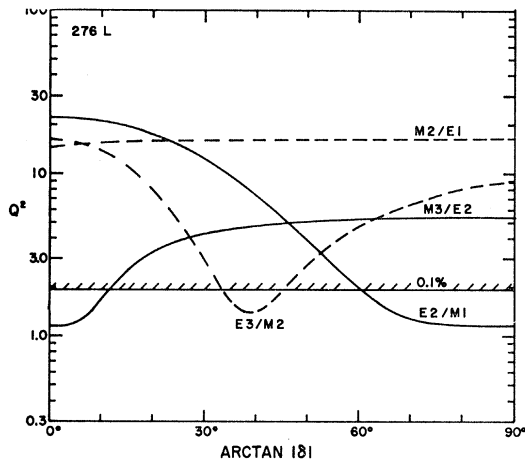


FIG. 13.  $Q^2$  versus  $\arctan |\delta|$  for the 276L line with  $\Delta B\rho/B\rho = 0.13\%$ .

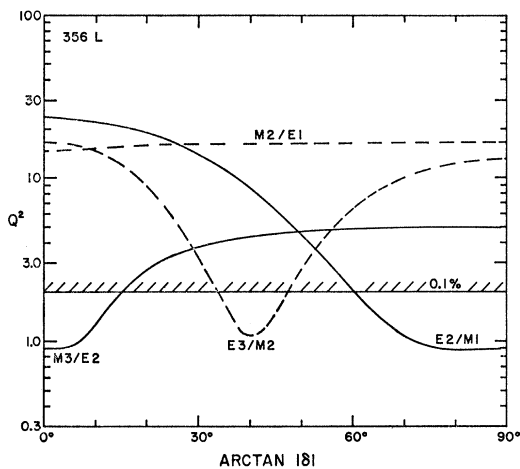


FIG. 14.  $Q^2$  versus  $\arctan |\delta|$  for the 356L line with  $\Delta B\rho/B\rho = 0.09\%$ .

### Discussion

The results of the  $K/L$  and  $L$ -subshell measurements presented in the previous section together with the recent  $\alpha_K$  measurements<sup>16-18</sup> restrict all the transitions, except the 223-keV transition, to connect states of the same parity. Consequently, all the levels populated by the electron-capture decay of  $Ba^{133}$  must have the same parity. From this fact it follows that the  $M2/E1$  mixing for the 223-keV transition, which is consistent with both the  $K/L$  and  $L$ -subshell measurements, is eliminated.

From the intensity limit  $I_{437K}/I_{356K} \leq 1 \times 10^{-4}$  obtained in the present work, one can make some weak arguments about the multipolarity of the 437-keV transition and the spin of the 437-keV level. Both spins  $\frac{1}{2}$  and  $\frac{3}{2}$  are consistent with the available  $356\gamma-81\gamma$  angular correlation data, although the value  $\frac{1}{2}$  is strongly preferred to fit the  $276\gamma-161\gamma$  angular correlation results. A spin  $\frac{3}{2}$  assignment would require that

the 437-keV transition be primarily  $E2$ , while spin  $\frac{1}{2}$  would require  $M3$ . Vartapetyan *et al.*<sup>34</sup> have recently reported an upper limit for the half-life of the 437-keV level,  $T_{1/2} \leq 1.5 \times 10^{-10}$  sec. If the half-life is taken to be exactly  $1.5 \times 10^{-10}$  sec, then the transition speeds relative to the Weisskopf estimates become  $|M(E2)|^2 \leq 7 \times 10^{-4}$  and  $|M(M3)|^2 \leq 2 \times 10^3$  for the 437-keV transition while  $|M(E2)|^2 = 12$  and  $|M(E2)|^2 = 5$  for the 356- and 276-keV transitions, respectively. Decreasing the lifetime by a factor of 1000 would make the reduced  $E2$  speed for the 437-keV transition more reasonable but would require excessive  $E2$  enhancement for the 276-keV and 356-keV transitions. Consequently, the generally accepted  $M3$  character for the 437-keV transition and  $\frac{1}{2}$  spin assignment for the 437-keV level are in better agreement with the present intensity measurements.

Assuming a spin sequence  $437(\frac{1}{2}^+)$ ,  $384(\frac{3}{2}^+)$ ,  $161(\frac{5}{2}^+)$ ,  $81(\frac{5}{2}^+)$ ,  $G.S.(\frac{7}{2}^+)$  we have compared the mixing ratios obtained from the present work to those resulting from previous measurements. The best values obtained from this comparison are presented in Fig. 17. In the following paragraphs the measurements pertinent to multipolarity mixings will be discussed for the basically pure  $E2$  transitions (276-, 356-, and 384-keV), the basically pure  $M1$  transitions (53-, 80-, 81-, 223-, and 303-keV), and the strongly mixed  $M1+E2$ , 161-keV transition.

The 276-keV transition is usually assumed to be of pure  $E2$  character. This assignment is consistent with the  $L$ -subshell measurements which give  $|\delta| \leq 0.084$  for  $M3$  admixture; similarly the  $K/L$  ratio yields  $|\delta| \leq 0.12$ .

The generally accepted  $E2$  character of the 356-keV transition is supported by the  $L$ -subshell measurements which give  $|\delta| \leq 0.03$  for  $M3$  admixture. The  $K/L$  measurement gives a somewhat broader limit,  $|\delta| \leq 0.2$ . The possibility of a mixture of 70%  $M1+30\%$   $E2$  suggested by Subba Rao<sup>25</sup> as a result of a measurement of the  $365-81K$  directional correlation is not supported by the present measurements. The  $L$ -subshell data would require an  $M1$  admixture of less than 6%. Recent measurements of the  $356\gamma-81K$ <sup>16,28</sup> and  $356K-81\gamma$ <sup>29,30,16</sup> directional correlations also are consistent with a predominantly  $E2$  character for the 356-keV transition.

While no  $L$ -subshell measurements were performed for the 384-keV transition, the measured  $K/L$  ratio shows that this transition is primarily  $E2$  with  $|\delta| \leq 0.45$  for  $M3$  admixture. The  $\alpha_K$  measurement of Thun *et al.*,<sup>16</sup>  $\alpha_K = 0.017 \pm 0.003$ , gives a more restrictive limit:  $|\delta| \leq 0.014$ . It should be mentioned that both of these measurements are also consistent with an  $M1$  admixture of 20% or less.

From our  $L_1/L_2$  measurement we find that the 53-keV transition is primarily  $M1$  with a small  $E2$  admixture such that  $|\delta| = 0.10 \pm 0.02$ . This value agrees with the result  $0.04 \leq \delta \leq 0.34$  obtained by Thun *et al.*<sup>16</sup>

from a measurement of the  $53K-384\gamma$  directional correlation. Thun *et al.*<sup>16</sup> also measured the  $K/L_1$  ratio of the 53-keV transition and obtained the limit  $|\delta| \leq 1.26$ .

The multipolarity mixing of the 81-keV transition has been extensively studied. Accurate  $L$ -subshell measurements by Brown *et al.*<sup>14</sup> and by Siegbahn *et al.*<sup>15</sup> are in agreement with an  $E2/M1$  mixture  $|\delta| = 0.164 \pm 0.006$ , while Erman and Sujkowski<sup>11</sup> give  $|\delta| = 0.181_{-0.026}^{+0.023}$  and  $|\delta| = 0.161_{-0.023}^{+0.020}$  from their  $L_1/L_2$  and  $L_1/L_3$  measurements, respectively. Our  $L$ -subshell data yield  $|\delta| = 0.173 \pm 0.004$ . (It should be noted that the error we quote for  $|\delta|$  is purely statistical and includes no error for the reference line shape used in the computer analysis.) There have been at least nine independent measurements of the  $356\gamma-81\gamma$  directional correlation.<sup>20-25,29,30,37</sup> A weighted average<sup>58</sup> of these results yields  $A_2 = 0.0373 \pm 0.0017$  and  $A_4 = -0.0020 \pm 0.0012$ . Assuming that the 356-keV transition is of pure  $E2$  multipolarity we get  $\delta = +0.153_{-0.002}^{+0.003}$ .<sup>59</sup>

Accurate information on the multipolarity mixing of the 80-keV transition is difficult to obtain. Our estimate  $L_1/L_2 \geq 8$  leads to an upper limit of  $|\delta| \leq 0.15$  for an  $E2$  admixture to a primarily  $M1$  transition. The  $K/L$  measurement resulting in  $|\delta| \leq 0.29$  substantiates this conclusion. Thun *et al.*<sup>16</sup> finds that  $|\delta| \leq 0.27, 0.28$ , and  $0.21$  from measurements of  $\alpha_K, K/L_1$ , and  $L_1/L_3$ , respectively. From a weighted average of the three reported measurements<sup>22-24</sup> of the 80-81  $\gamma-\gamma$  directional correlation we get  $A_2 = 0.0380 \pm 0.0035$  and  $A_4 = -0.0030 \pm 0.0026$ . Combining this result with the value  $\delta_{81} = 0.153_{-0.002}^{+0.003}$  we get  $\delta_{80} = 0.13_{-0.07}^{+0.08}$ .

Substantial agreement for the  $E2/M1$  mixing of the 223-keV transition is obtained from the  $L$ -subshell measurement resulting in  $|\delta| = 0.40_{-0.18}^{+0.14}$  and the  $K/L$  measurement leading to  $|\delta| \leq 0.4$ . The only other previous information on the multipolarity of this transition is the measurement by Nieschmidt *et al.*,<sup>13</sup>  $K/(L+M) = 7.4 \pm 0.07$ , leading to an assignment of  $E0, M1$ , or  $E0+M1$  mixture.

The 303-keV transition appears to be quite pure  $M1$  transition with an  $E2$  admixture  $|\delta| = 0.11_{-0.11}^{+0.07}$  from the  $L$ -subshell measurement or  $|\delta| = 0.6_{-0.6}^{+0.5}$  from the  $K/L$  ratio. The three reported measurements<sup>22-24</sup> of the 303-81 directional correlation are in substantial agreement. Using a weighted averaging process on the correlation data we calculate that  $A_2 = -0.0253$

$\pm 0.0026$  and  $A_4 = +0.0028 \pm 0.0021$ . Again with  $\delta_{81} = 0.153_{-0.002}^{+0.003}$ , the multipolarity mixing becomes  $\delta_{303} = 0.006_{-0.021}^{+0.022}$ .

The 161-keV transition is probably the most interesting transition in the decay scheme. Four measurements of the 276-161 directional correlation have been reported.<sup>20,23,29,38</sup> A weighted average of these measurements gives  $A_2 = -0.435 \pm 0.007$  and  $A_4 = -0.025 \pm 0.009$ . Assuming a spin sequence  $\frac{1}{2}(E2)\frac{5}{2}(\delta)\frac{7}{2}$ , we obtain two solutions  $\delta_1 = -0.63 \pm 0.02$  and  $\delta_2 = -2.21 \pm 0.10$  from  $A_2$ . Furthermore, only  $\delta_1$  is consistent with the value of  $A_4$ ; more than twice the above quoted error limit on  $A_4$  must be invoked to get agreement with  $\delta_2$ .

The angular distribution of 161-keV  $\gamma$  rays produced in the coulomb excitation of  $Cs^{133}$  by 16.1-MeV nitrogen ions has been investigated recently by Alkhazov *et al.*<sup>31</sup> The two solutions corresponding to a spin sequence  $\frac{7}{2}(E2)\frac{5}{2}(\delta)\frac{7}{2}$  are:  $\delta_1 = -0.46_{-0.11}^{+0.08}$  and  $\delta_2 = -3.2_{-1.1}^{+0.8}$ . These values are in reasonable agreement with the 276-161 directional correlation results. It is, perhaps, worth mentioning that the 276-161  $\gamma-\gamma$  directional correlation and the angular distribution of the 161-keV gamma rays following Coulomb excitation give a measure of the same quantity, namely  $A_2^{(2)}(\frac{5}{2} \rightarrow \frac{7}{2}, \delta_{161}) = (0.13363 - 1.38873\delta_{161} + 0.32453\delta_{161}^2)/(1 + \delta_{161}^2)$ . Consequently, two such measurements could not be used to decide between the two possible mixing ratios  $\delta_1$  and  $\delta_2$ .

The value of  $\epsilon B(E2)$  was also measured by Alkhazov *et al.* Utilizing the values  $\alpha_T(161) = 0.5$ ,  $\Gamma_\gamma(80)/\Gamma_\gamma(161) = 8$ , and  $\tau(161) \leq 7 \times 10^{-10}$  sec., they have calculated  $B(E2)\uparrow = 0.13 \pm 0.04$ ,  $\tau(E2) = (4.5 \pm 1.5) \times 10^{-9}$  sec. and consequently  $|\delta| \leq 0.55$ . This argument led them to choose the smaller of the two possible values of the mixing obtained from the angular-distribution measurements.

The present  $L$ -subshell measurements appear to be in serious contradiction to the directional correlation results. Three independent measurements of the  $L$ -line intensities of this transition have been made as indicated in Table IV. A weighted average of these results gives  $|\delta| = 1.06$  with a weighted internal error of 0.06 or an external error of 0.05. Examination of the  $Q^2$  versus  $|\delta|$  plot for this transition (Fig. 9) also shows that the mixing ratio  $|\delta| = 0.63$  obtained from the correlation work corresponds to a  $Q^2$  value in excess of the 0.1% confidence limit. The existence of some systematic effects appears unlikely because of the good consistency between the separate measurements and the general agreement of the other  $L$ -subshell measurements with previous measurements. The possibility that this measurement might be particularly sensitive to the  $L$ -subshell conversion coefficients interpolated from the tables of Sliv and Band<sup>55</sup> was tested by a  $\pm 5\%$  variation of the conversion coefficients utilized in the computer analysis of the  $L$ -subshell data. The extreme variations for  $|\delta|$  obtained in this way would result in an error twice as large as quoted above. This

<sup>58</sup> For calculating a weighted average of  $A_2$  we utilize the weighting factor  $\omega_i = (\Delta A_{2i})^{-2} / \sum_j (\Delta A_{2j})^{-2}$  to get  $\langle A_2 \rangle_{av} = \sum_i \omega_i A_{2i}$ . The error quoted is the larger of the two quantities  $\sigma_{int}^2 = \sum_i (\Delta A_{2i})^{-2}$  and  $\sigma_{ext}^2 = (N-1)^{-1} \sum_i \omega_i [A_{2i} - \langle A_2 \rangle_{av}]^2$ , where  $N$  is the total number of measurements. The quantity  $A_4$  is treated in the same manner.

<sup>59</sup> In the analysis of angular correlations we utilize the phase convention of A. J. Ferguson in *Angular Correlations Methods in Gamma-Ray Spectroscopy* (Interscience Publishers, Inc., New York, 1965), p. 5 rather than the more familiar convention of L. C. Biedenharn and M. E. Rose, *Rev. Mod. Phys.* **25**, 729 (1953).

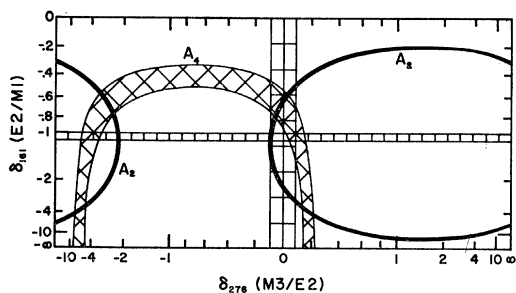


FIG. 15. Contour diagram representation of the 276-161  $\gamma$ - $\gamma$  angular correlation for a spin sequence  $\frac{1}{2}^+ - \frac{5}{2}^- - \frac{7}{2}^-$ . Contours of  $A_2 = -0.435 \pm 0.007$  and  $A_4 = -0.025 \pm 0.009$  are shown as well as the limitations on  $\delta_{161}$  and  $\delta_{276}$  obtained from the  $L$ -subshell measurements.

extension of the error limits would still not easily explain the discrepancy. The use of the conversion coefficients of Rose<sup>60</sup> also does not affect the situation in a significant manner.

Another possible explanation for this situation can be seen by examining Fig. 15 which shows a contour diagram of  $A_2$  and  $A_4$  from the  $\gamma$ - $\gamma$  angular-correlation measurements together with the multipolarity mixings obtained from the  $L$ -subshell measurements. Agreement between the  $L$ -subshell values and the  $A_2$  value is obtained if one allows an  $M3/E2$  mixture for the 276-keV transition of  $-0.11 \leq \delta \leq -0.07$ . The value of  $A_4$  would have to be changed by more than twice the error limit of 0.09 in order to agree with this solution. Furthermore, the estimated half-life of the 437-keV level  $T_{1/2} \leq 1.5 \times 10^{-10}$  sec<sup>34,35</sup> would indicate that such an admixture would require an  $M3$  enhancement of  $10^5$ - $10^6$  relative to the single-particle estimate. An enhancement of this magnitude appears unlikely in view of the existing information on  $M3$  strengths.

Using the multipolarity mixing  $\delta^2 = 0.4$  obtained from the double correlation data together with the measured half-life<sup>38</sup>  $T_{1/2} = (0.85 \pm 0.16) \times 10^{-10}$  sec for the 161-keV level, one can estimate that the  $M1$  portion of the 161-keV transition is hindered by roughly a factor of 300. Such a hindrance is of the same order of magnitude as found for the 81-keV transition for which the possibility of penetration effects on the conversion electrons has been extensively examined. These facts would suggest that the possibility of penetration effects should also be examined in the present case. To this end we shall assume that the penetration effects for the  $M1$  conversion coefficients can be written in the usual way<sup>61,62</sup>

$$\beta_i(\lambda) \approx \beta_i(1) [1 - (\lambda - 1)C_i]^2. \quad (5)$$

Here  $i = K, L_1, L_2, L_3$ ,  $\beta_i(1) = \beta_i(\text{Sliv})$ ,  $\lambda =$  ratio of conversion matrix element to gamma-ray matrix

<sup>60</sup> M. E. Rose, *Internal Conversion Coefficients* (North-Holland Publishing Company, Amsterdam, 1958).

<sup>61</sup> E. L. Church and J. Weneser, *Phys. Rev.* **104**, 1382 (1956).

<sup>62</sup> E. L. Church and J. Weneser, *Bull. Am. Phys. Soc.* **7**, 490 (1962); text of talk distributed as Brookhaven National Laboratory Report No. BNL-6647 (unpublished).

element,  $C_K = 0.0080$ ,<sup>61</sup>  $C_{L_1} = 0.0098$ .<sup>63</sup> One can also safely assume that in the present case the penetration effects for the  $L_3$  shell are negligible as they were for the case of the 279-keV transition in  $\text{Tl}^{203}$ .<sup>62</sup> Hence, with the coefficients presently available one can examine the penetration effects for  $\alpha_K$ ,  $K/L_3$ , and  $L_1/L_3$ .

In order to obtain the  $K/L_3$  and  $L_1/L_3$  ratios the existing stripping program used for  $L$ -subshell analysis was modified to give  $L_1/L_2/L_3$  relative intensities. Using a weighted average of the resulting  $L$ -subshell ratios together with the measured  $K/L$  ratio, we obtain  $K/L_3 = 20.8 \pm 2.4$  and  $L_1/L_3 = 2.17 \pm 0.13$ . Figure 16 shows a contour diagram in the  $\lambda$ - $|\delta_{161}|$  plane for the quantities  $L_1/L_3$  and  $K/L_3$  in addition to the two values of  $|\delta_{161}|$  obtained from the 276-161  $\gamma$ - $\gamma$  angular correlation. Also shown in Fig. 16 is a contour labeled  $B(E2)$  representing the quantity

$$\begin{aligned} & [\tau \epsilon B(E2) \times 1.72 \times 10^9]^{1/2} \\ &= \frac{\delta(1 + \delta^2)^{1/2}}{1 + \delta^2 + I[\beta_K(\lambda) + \delta^2 \alpha_K(E2)]} \\ &= 0.0457 \pm 0.0082. \end{aligned} \quad (6)$$

Here

$$\begin{aligned} I &= \left\{ \frac{1 + K/(L+M)}{K/(L+M)} \right\}_{161} + \frac{I_T(80)}{I_K(161)} \\ &= 46.0 \pm 9.6, \end{aligned} \quad (7)$$

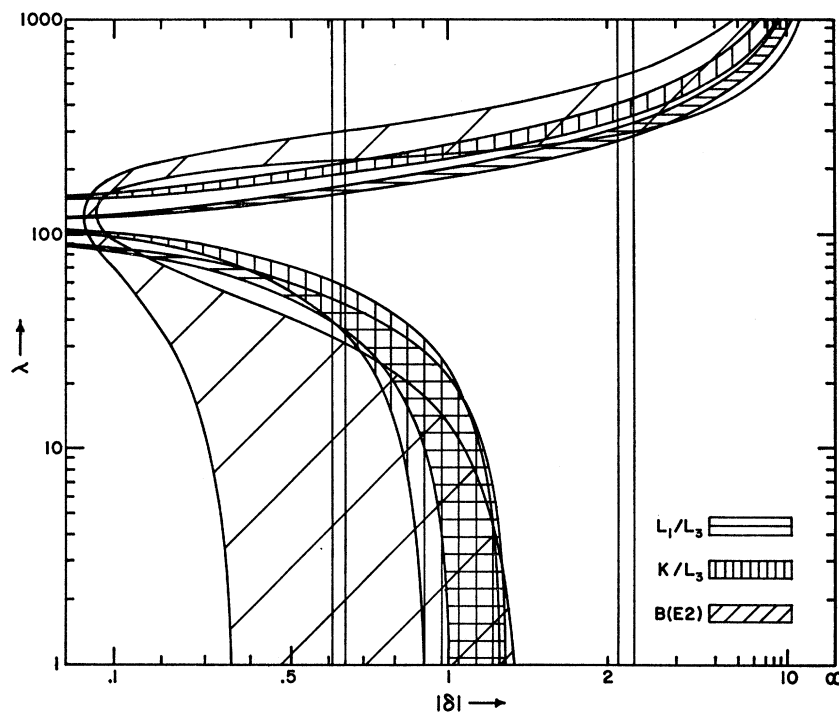
with  $\epsilon B(E2) = 0.0096$  ( $e^2 \times 10^{-48}$  cm<sup>4</sup>)<sup>31</sup> and  $\tau = 1.26 \times 10^{-10}$  sec.<sup>33</sup>

Clearly the bands in Fig. 16 corresponding to  $K/L_3$  and  $L_1/L_3$  are the most restrictive and give a solution in the region  $|\delta| = 0.63 \pm 0.02$  and  $\lambda_1 = 45 \pm 10$ . Unfortunately the contours for  $K/L_3$  and  $L_1/L_3$  appear to exhibit very similar behavior as a function of  $\lambda$  and  $|\delta|$ . If one includes the generally accepted  $\pm 10\%$  accuracy of the values for  $C_K$  and  $C_{L_1}$ ,<sup>61,62</sup> two other regions of solution must also be considered, namely:  $|\delta| = 0.63 \pm 0.02$  with  $\lambda_2 = 180 \pm 20$  or  $|\delta| = 2.2 \pm 0.1$  with  $\lambda_3 = 350 \pm 50$ . The last solution is not consistent with the  $A_4$  value obtained from the 276-161  $\gamma$ - $\gamma$  directional correlations. The contour resulting from the  $B(E2)$  measurement does not clarify the picture further; it clearly agrees with solution three and possibly with solution one, while even the second solution can not be completely eliminated.

There exist two recent measurements of the  $K$ -conversion coefficient for the 161-keV transition. The measurement of Thun *et al.*,<sup>16</sup>  $\alpha_K = 0.39 \pm 0.13$ , is consistent only with solution three; while the measurement of Szichman *et al.*,<sup>17</sup>  $\alpha_K = 0.18 \pm 0.03$ , agrees with solutions one and two. Clearly a remeasurement of  $\alpha_K(161)$  would be useful for deciding which value of  $\lambda$  best fits the data for this transition. Another measurement of value for this purpose would be the 276 $\gamma$ -161 $e_K$

<sup>63</sup> A. S. Reiner, *Nucl. Phys.* **5**, 544 (1958).

FIG. 16. Contour diagram representation of penetration effects for the 161-keV transition in  $\text{Cs}^{133}$ . Contours of  $K/L_3$ ,  $L_1/L_3$ , and  $[\tau\epsilon B(E2) \times 1.72 \times 10^{+9}]^{1/2}$  are shown. The latter quantity is indicated by the label " $B(E2)$ ". The two vertical bands represent the two possible values of  $\delta_{161}$  obtained from the 276-161 double correlation assuming a pure  $E2$  276-keV transition and a spin sequence  $\frac{1}{2}^+ - \frac{3}{2}^- - \frac{1}{2}^-$ .



or  $276e_K - 161\gamma$  correlation. Meder *et al.*<sup>29</sup> have recently reported such a  $\gamma - e_K$  measurement which indeed does not agree with the  $\gamma - \gamma$  results if  $\lambda_{161} = 1$ . Penetration effects for this measurement are presently being investigated.<sup>64</sup>

A review of the literature indicates that, with the exception of the careful work reported by Bodenstedt *et al.*,<sup>20</sup> the bulk of the double-correlation measurements have not been exhaustively examined to ascertain the possible existence of other spin sequences consistent with the data. It would seem to be worthwhile to reexamine the various directional-correlation data in light of the additional information available on the transition multipolarity mixings as a result of the present  $L$ -subshell and recent  $K$ -conversion-coefficient measurements. The establishment of nuclear-spin assignments by the use of such information, which is primarily independent of nuclear models, is preferable to the use of the weaker arguments based upon shell model predictions, electron-capture intensities, and  $\gamma$ -ray intensities.<sup>65</sup> To this end the available data on the  $\gamma - \gamma$  directional correlations, 356-81, 276-161, 80-81, and 303-81, together with the  $L$ -subshell and  $\alpha_K$  measurements were used to test the following spin possibilities:  $437(\frac{1}{2}, \frac{3}{2}, \frac{5}{2})$ ,  $384(\frac{1}{2}, \frac{3}{2}, \frac{5}{2})$ ,  $161(\frac{3}{2}, \frac{5}{2}, \frac{7}{2}, \frac{9}{2})$ ,  $81(\frac{5}{2}, \frac{7}{2}, \frac{9}{2})$ ,  $G.S.(\frac{7}{2})$ . The correlation data were analyzed by calculating contour plots of  $A_2$  and  $A_4$  in the  $\arctan \delta_p - \arctan \delta_s$  plane for each spin sequence. Here  $\delta_p$  and  $\delta_s$

refer to the multipolarity mixing ratio for the primary and secondary radiations, respectively. Only mixings of quadrupole/dipole, octupole/quadrupole, and pure octupole were considered. While several solutions were found for each double cascade (in particular the 80-81 cascade agrees with all the spin sequences investigated except  $\frac{3}{2}^+ - \frac{7}{2}^- - \frac{7}{2}^-$ ), no new spin sequence was found to agree with all the available information on multipolarities and correlations.

A special comment is in order on the possibility of an  $E0$  admixture consistent with the  $L$ -subshell and  $K/L$  measurements for the 303- and 223-keV transitions. The data for the 303-81 directional correlation is consistent with a spin sequence  $\frac{5}{2}^+ - \frac{5}{2}^- - \frac{7}{2}^-$  together with the accepted value of  $\delta_{81} = 0.153_{-0.002}^{+0.003}$  and the values  $\delta_{303} = -0.92_{-0.13}^{+0.09}$  or  $-5.0_{-2.0}^{+1.3}$ . Both of these solutions for  $\delta_{303}$  would agree with the value  $|\delta|_{303} \geq 0.36$  which is found from the  $L$ -subshell measurements if one allows an  $E0$  admixture  $0.2 \leq \epsilon^2 \leq 0.5$  as discussed previously. However, the measurements of  $\alpha_K(303)$ <sup>16,18</sup> restrict any  $E0$  admixture to be  $\epsilon^2 \leq 0.03$ . Hence, the 384-keV level cannot have a spin of  $\frac{5}{2}$ .

An  $E0$  admixture for the 223-keV transition would require that the 384- and 161-keV levels have the same spin, namely either  $\frac{3}{2}$  or  $\frac{5}{2}$ . The spin  $\frac{3}{2}$  is ruled out by the 276-161  $\gamma - \gamma$  double correlation; while the value  $\frac{5}{2}$  is in disagreement with the  $\alpha_K(303)$  measurements<sup>16,18</sup> and present  $L$ -subshell measurements for the 303-keV transition as shown in the previous paragraph. Consequently an  $E0$  admixture for the 223-keV transition can also be ruled out.

<sup>64</sup> M. R. Meder (private communication).

<sup>65</sup> K. Way, in *Nuclear Spin Parity Assignments*, edited by N. B. Gove and R. L. Robinson (Academic Press Inc., New York, 1966), p. 1-4.

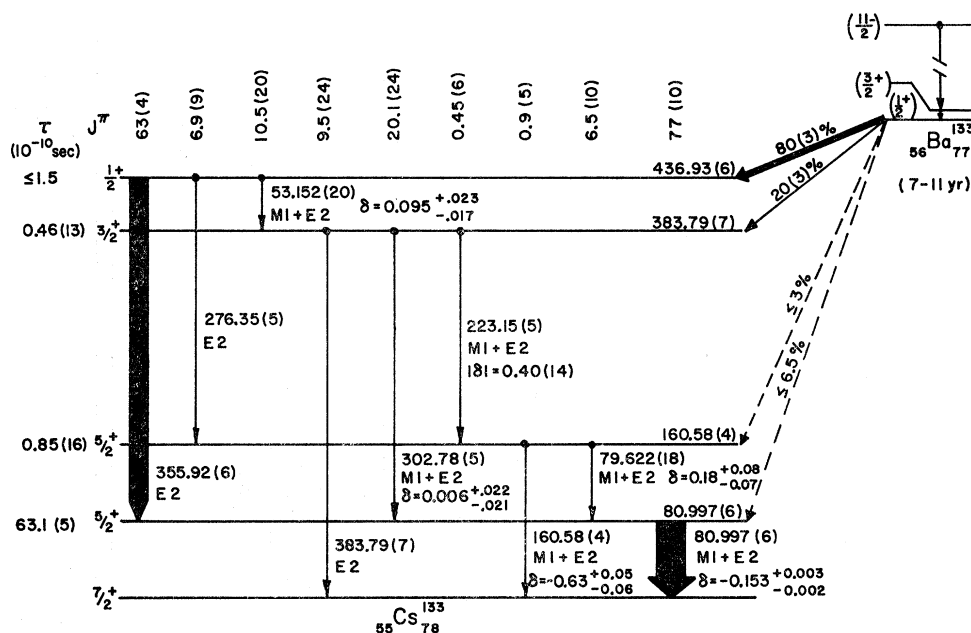


FIG. 17.  $\text{Cs}^{133}$  level scheme resulting from the decay of  $\text{Ba}^{133}$ . All energies are in keV and total transition intensities in percent of decays of  $\text{Ba}^{133}$ .

The present internal-conversion-electron intensity measurements combined with reported  $\alpha_K$  measurements have been utilized to calculate the total electromagnetic transition probabilities for  $\text{Cs}^{133}$  and the electron-capture branching from  $\text{Ba}^{133}$ . The results are shown in Fig. 17 where the electromagnetic transition intensities are normalized to 100 decays of  $\text{Ba}^{133}$ . The large errors quoted for the 161-keV transition reflect the uncertainty in the value of  $\alpha_K(161)$ . In spite of the uncertainty for the intensity of this transition, it is clear from our measurements that the electron-capture feeding to the 161-keV level must be less than 3% in contrast to the 13% feeding to this level reported by Stewart and Lu.<sup>9</sup> This result removes one of the remaining arguments for a spin assignment of  $3/2$  to the 161-keV level.

The  $\gamma$ -ray branching ratios for the 384-, 303-, and 223-keV transitions relative to the total electromagnetic transition decay probability for the 384-keV level have also been computed to be  $\epsilon_\gamma(384) = 0.31 \pm 0.06$ ,  $\epsilon_\gamma(303) = 0.64 \pm 0.06$ , and  $\epsilon_\gamma(223) = 0.014 \pm 0.02$ . Utilizing the value of  $\epsilon_\gamma(384)$  together with the measurement of  $B(E2) = 0.023 \pm 0.005$  in units of  $e^2 \times 10^{-48} \text{ cm}^4$  for the

384-keV transition by Alkhozov *et al.*,<sup>32</sup> we obtain  $T_{1/2} = 0.46 \pm 0.13 \times 10^{-10}$  sec for the half-life of the 384-keV level.<sup>66</sup> The speeds for these three transitions relative to the Weisskopf estimates have also been calculated. The computations indicate that the  $E2$  components of the 384-, 303-, and 223-keV transitions are enhanced by factors of 11,  $\leq 3.4$  and 1.0, respectively. The  $M1$  components of the 303- and 223-keV transitions are hindered by factors of 97 and 1950, respectively. The reduced transition speeds for the other transition seen in the decay of  $\text{Ba}^{133}$  have been presented by numerous authors (see Thun *et al.*<sup>16</sup> for the latest estimates). In general both the  $E2$  and  $M1$  reduced speeds are typical for transitions in this mass region.

#### ACKNOWLEDGMENTS

We wish to thank Dr. D. D. Watson and Lt. A. K. Hyder for their advice relative to the spectrum stripping programs utilized in this work and to Dr. G. I. Harris for a critical reading of the manuscript.

<sup>66</sup> The calculated mean life for the 384-keV transition given in Table I of Ref. 32 appears to be a misprint.