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# GAMMA-GAMMA DIRECTIONAL CORRELATION MEASUREMENTS IN THE DECAY OF Ba133

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Abstract: The gamma rays following the electron capture decay of Ba138 have been studied using coincidence and directional correlation techniques. The existence of gamma rays with energies of 162 keV and 222 keV has been ascertained. The correlation functions for the 356 keV-82 keV, 302 keV-82 keV, 80 keV-82 keV and 276 keV-162 keV cascades are found to be, respectively:  $W(\theta) = 1 + (0.0331 \pm 0.0017) P_2(\cos \theta) + (0.0045 \pm 0.0033) P_4(\cos \theta),$ 

 $W(\theta) = 1 - (0.0238 \pm 0.0060) P_2(\cos \theta) + (0.0061 \pm 0.0089) P_4(\cos \theta),$ 

 $W(\theta) = 1 + (0.0319) \pm 0.0045) P_2(\cos \theta) + (0.005 \pm 0.009) P_4(\cos \theta),$ 

 $W(\theta) = 1 - (0.328 \pm 0.009) P_2(\cos \theta) - (0.067 \pm 0.016) P_4(\cos \theta).$ 

These results support the spin sequence of  $\frac{7}{4}$ ,  $\frac{5}{4}$ ,  $\frac{5}{4}$ ,  $\frac{5}{4}$ , and  $\frac{1}{4}$ , for the ground state, 82, 162, 384 and 438 keV levels of Cs133. The measured mixing ratios of the following gamma rays are: 80 keV: $(63\pm9)\%$  E2+ $(37\pm9)\%$  M1; 82 keV:  $(97.6\pm0.1)\%$  M1+ $(2.4\pm0.1)\%$  E2; 162 keV:  $(95.1 \pm 0.6)\%$  E2+ $(4.9 \pm 0.6)\%$  M1; 276 keV: pure E2; 302 keV:  $(99.7 \pm 0.1)\%$  M1+ $(0.3 \pm 0.1)\%$ E2; 356 keV: pure E2.

RADIOACTIVITY Ba188 [from Oak Ridge]; measured  $\gamma\gamma$ -coin,  $\gamma\gamma(\theta)$ . Cs<sup>133</sup> deduced levels, J,  $\pi$ ,  $\delta(E2/M1)$ .

## 1. Introduction

The nuclear levels of Cs<sup>133</sup> have been studied by many authors <sup>1-13</sup>). Most of the recent investigators 7,8,14) are in agreement concerning the salient features of the electron capture decay scheme of 7.2 y Ba<sup>133</sup> as shown in fig. 1. Excited levels of Cs<sup>133</sup> at 82, 162, 384 and 438 keV have been established. The majority of authors are also in accord concerning the existence of all gamma ray transitions shown in fig. 1, except the two of energies 162 keV and 222 keV. These two gamma rays have been observed by Stewart and Lu 8), but not by Ramaswamy et al. 11). Through the measurement of relative gamma ray intensities, Stewart and Lu<sup>8</sup>) also deduced an additional electron capture transition from the ground state of Ba<sup>133</sup> leading directly to the 162 keV level; whereas other authors observed only the two transitions leading to the 384 and the 438 keV levels. Although the spins and parities of the ground state, 82, 384 and 438 keV levels of Cs<sup>133</sup> are fairly certain to be  $\frac{7}{2}$ ,  $\frac{5}{2}$ ,  $\frac{3}{2}$  and  $\frac{1}{2}$  respectively, the spin of the 162 keV level has been a point of dissent among authors. Bodenstedt et al. 7) determined the spin of this level to be \(\frac{5}{2}\) through directional

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correlation measurement of the 276 keV-162 keV cascade; although they could not determine the mixing ratio of the 162 keV gamma ray from their results. Ramaswamy et al.  $^{11,12}$ ) also determined the spin of this level to be  $\frac{5}{2}$  based on the absence of an electron capture transition leading to this level from the ground state of Ba<sup>133</sup> ( $\frac{1}{2}$ ). However, since Stewart and Lu  $^8$ ) did find an electron capture transition leading to this level, their interpretation of this branch as an allowed transition (log ft = 9) requires the spin to be  $\frac{3}{2}$ . Recently, the directional correlation measurement of Arya  $^{13}$ ) on the 80 keV-82 keV cascade also assigns a spin of  $\frac{3}{2}$  to this level. It is the purpose of the present investigation to resolve some of these uncertainties.

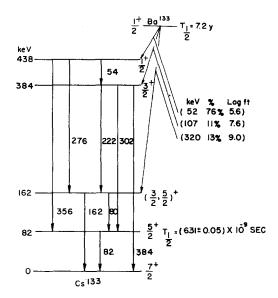


Fig. 1. Decay scheme of Ba<sup>133</sup>.

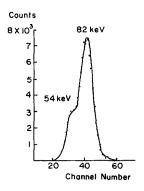
## 2. Experimental Procedure

Ba<sup>133</sup> in the form of HCl solution was obtained from Oak Ridge. The liquid source, upon dilution, was injected into a thin-walled cylindrical lucite container of 4 mm diameter for both coincidence and directional correlation measurements. Two 5.1 cm by 5.1 cm NaI(Tl) crystals mounted on RCA 6342A phototubes were used as detectors, with lateral lead shieldings to eliminate counter-to-counter scatterings. A fast-slow coincidence circuit with a resolving time of about 40 ns was employed in all cases; and energy selection was provided by two differential analysers. For coincidence measurements, the pulses coincident with the selected energy range were fed through a linear gate to be recorded on a 256-channel analyser. For each coincidence measurement, a corresponding accidental coincidence measurement for the same duration was also taken and subtracted in order to obtain the true coin-

cidence spectrum. In all directional correlation measurements, the source to detector distance was 7 cm; and data were taken at every 15° in a double-quadrant sequence. The instruments were calibrated after each run to insure their stability. The true coincidence rate was furthermore divided by the product of the single counting rates to correct for possible electronic drifts. After making a least-square fit of the correlation data 15), the expansion coefficients were normalized and corrected for finite angular resolution 16).

## 3. Coincidence Results

As a preliminary preparation for directional correlation measurements, a series of coincidence spectra was collected. All results confirm the decay scheme of fig. 1. A few of the coincidence spectra of special interest will be presented here.



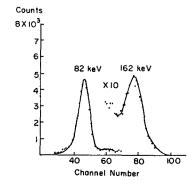


Fig. 2. Spectrum of gamma rays in coincidence with energies greater than 374 keV.

Fig. 3. Spectrum of gamma rays in coincidence with the energy range 264 to 284 keV.

Fig. 2 shows the spectrum of gamma rays which are coincident with energies greater than 374 keV. Since the selecting discriminator inevitably accepts some portion of the 356 keV photopeak, one notices a rather pronounced 82 keV peak in the coincidence spectrum. However, it is also clear that there is an enhancement at 54 keV which is much more pronounced than the normal escape peak of the 82 keV gamma ray. The existence of this gamma ray at 54 keV in coincidence with energies higher than 374 keV lends support to the decay scheme of fig. 1.

Fig. 3 shows the spectrum of gamma rays in coincidence with the energy range of 264 to 284 keV. The peaks at 80 keV and 162 keV are clearly evident. This spectrum demonstrates not only the existence of the 162 keV gamma ray, but also its coincidence relationship with the 276 keV gamma ray. Since the selected energy range also includes the Compton continuum of higher energy gamma rays, a portion of the area underneath the 80 keV peak is due to the contribution of the 82 keV gamma ray which is coincident with these high energy gamma rays as well as with the 276 keV itself.

The coincidence spectrum of the energy range, 148 to 172 keV, is shown in fig. 4. The 276 keV gamma ray, whose intensity is so low that its photopeak is almost completely masked by the 356 and 302 keV gamma rays in the single spectrum, now stands out very prominently. Also noteworthy is the weak, but definite peak at 222 keV. The existence of this gamma ray was previously considered questionable. The peak at 162 keV is coincident with the Compton continuum of the 276 keV gamma ray, which is also being accepted in the energy selector.

The single spectrum of Ba<sup>133</sup> as well as other coincidence spectra are well known in the literature <sup>1-13</sup>). Therefore, in order to avoid repetition, they will not be presented here. In general, it can be said that all coincidence measurements completely supports the decay scheme of fig. 1.

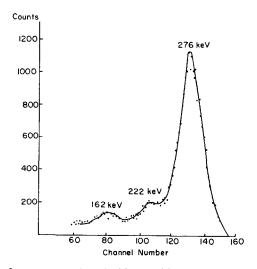


Fig. 4. Spectrum of gamma rays in coincidence with the energy range 148 to 172 keV.

## 4. Directional Correlation Results

Since the 82 keV level has a half-life <sup>7</sup>) of  $6.31\pm0.05$  ns, possible attenuation of the directional correlations involving this level as an intermediate state may occur. Several authors have investigated this possibility. Clikeman and Stewart <sup>9</sup>) used both solid BaCl<sub>2</sub> and liquid sources for the correlation of the 356 keV-82 keV cascade. They found no difference in anisotropy within experimental error. Bodenstedt et al. <sup>7</sup>) used a liquid source to measure the correlation of the same cascade, and observed the anisotropy as a function of the time delay between the two gamma rays. Their results also showed no observable attenuation. Based on the conclusion reached by these authors, as well as the fact that the source used in the present experiment is in the form of dilute HCl solution, no perturbation of the directional correlations involving the 82 keV level as the intermediate state is expected. Since the half-life

of the 162 keV level is shorter ( $\leq 0.5$  ns, see ref. <sup>7</sup>)) than that of the 82 keV level, it is also assumed that no corrections for the perturbations by extranuclear fields are necessary for the 276 keV-162 keV directional correlation function.

#### 4.1. THE 356 keV-82 keV CASCADE

One differential analyser was set to accept the 356 keV photopeak, ("A" of fig. 5), being careful to avoid contribution of the 302 keV line; while the other analyser accepted the photopeak of the 82 keV gamma ray. About  $2.7 \times 10^5$  true coincidences were accumulated at each angle. The normalized expansion coefficients, after correction for finite resolution, are:

$$A_2 = 0.0331 \pm 0.0017, \qquad A_4 = 0.0045 \pm 0.0033.$$

Although the error in  $A_4$  is large enough to cast doubt upon its sign, it is sufficiently small to restrict the magnitude of the absolute value of  $A_4$ .

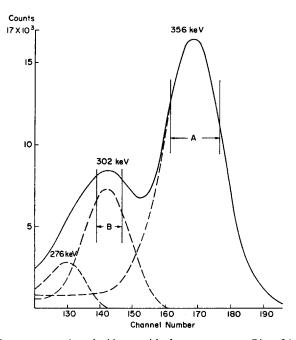


Fig. 5. Spectrum of gamma rays in coincidence with the energy range 74 to 94 keV. The complex peaks are resolved with the mono-energetic pulse height shapes of Hg<sup>808</sup> (279 keV) and Sn<sup>118</sup> (392 keV).

# 4.2. THE 302 keV-82 keV CASCADE

Since interference from other cascades, notably that of 356 keV-82 keV, was unavoidable, the spectrum of gamma rays in coincidence with 82 keV was taken. The photopeaks at 356, 302 and 276 keV in this spectrum were resolved with the help of the mono-energetic pulse height shapes of Hg<sup>203</sup> (279 keV) and Sn<sup>113</sup> (392

keV), as shown in fig. 5. The differential analyser accepting the 302 keV gamma ray was set at position "B" of fig. 5, with a negligible amount of overlap with the 276 keV line. The other discriminator was set to accept the 82 keV photopeak. The interference from the 356 keV-82 keV cascade was found to be 14%.

A total of about  $7.3 \times 10^4$  true coincidences were accumulated at each angle, and the normalized expansion coefficients, after correcting for finite resolution, are:

$$A_2 = -0.0159 \pm 0.0052, \quad A_4 = 0.0059 \pm 0.0077.$$

The revised coefficients, after correction for the interference of the 356 keV-82 keV cascade, are:

$$A_2 = -0.0238 \pm 0.0060, \quad A_4 = 0.0061 \pm 0.0089.$$

#### 4.3. THE 80 keV-82 keV CASCADE

For this cascade, each discriminator was set to accept both of the photopeaks. Using the method described in subsect. 4.2, the interference from the 356 keV-82 keV cascade was found to be 10%, and that of the 302 keV-82 keV cascade, 3%. The true coincidences accumulated at each angle were about  $4.7 \times 10^4$ , and the final normalized coefficients, corrected for both angular resolution and interferences, are:

$$A_2 = 0.0319 \pm 0.0045, \quad A_4 = 0.005 \pm 0.009.$$

#### 4.4. THE 276 keV-162 keV CASCADE

Although the coincidence rate for this cascade is quite small compared to the other cascades, the interferences from other cascades are not present; and the anisotropy is considerably more pronounced than the others. The differential analysers were set to accept the energy ranges of 140 to 180 keV and 254 to 294 keV respectively. A total of about  $2 \times 10^4$  true coincidences were accumulated at each angle, and the normalized coefficients, after geometry corrections, are:

$$A_2 = -0.328 \pm 0.009$$
,  $A_4 = -0.067 \pm 0.016$ .

## 5. Interpretation and Discussion

#### 5.1. THE 356 keV-82 keV CASCADE

The spins of the ground state of  $Cs^{133}$  and  $Ba^{133}$  are  $^{10,17}$ )  $\frac{7}{2}^+$  and  $\frac{1}{2}^+$  respectively, in agreement with shell model predictions of  $g_{\frac{1}{2}}$  and  $s_{\frac{1}{2}}$ . Judging from the allowed nature of the electron capture transition from the ground state of  $Ba^{133}$  ( $\frac{1}{2}^+$ ) to the 438 keV level, ( $\log ft = 5.6^{-14}$ );  $\log ft = 6.1^{-12}$ ), and the fact that the 438 keV level has not been reached by Coulomb excitation 5), the spin and parity of the 438 keV level is assumed to be  $\frac{1}{2}^+$ . The 82 keV level, on the other hand, has been reached by Coulomb excitation 5). It is populated from the ground state  $^{14}$ ) of  $Xe^{133}$  ( $\frac{3}{2}^+$ ) through an allowed  $\beta$  transition ( $\log ft = 5.6$  see ref.  $^{14}$ )); but not populated by an electron capture transition from the ground state of  $Ba^{133}$  ( $\frac{1}{2}^+$ ). All these facts, in

addition to the predominantly M1 nature of the 82 keV gamma ray  $^9$ ) as deduced from its internal conversion coefficient, combine to assign uniquely a spin and parity of  $\frac{5}{2}$  to the 82 keV level.

The spin sequence for the 356 keV-82 keV cascade is therefore  $\frac{1}{2}^+(Q)^{\frac{5}{2}^+}(D,Q)^{\frac{7}{2}^+}$ . The theoretical  $A_2^{(1)}$  and  $A_4^{(1)}$  obtained from the table of F coefficients <sup>18</sup>) are:

$$A_2^{(1)} = -0.5345, \quad A_4^{(1)} = -0.6172.$$

Knowing  $A_2 = 0.0331 \pm 0.0017$  and  $A_4 = 0.0045 \pm 0.0033$ , one obtains for the 82 keV gamma ray:

$$A_2^{(2)\text{exp}} = -0.0619 \pm 0.0032, \quad A_4^{(2)\text{exp}} = -0.0073 \pm 0.0053.$$

Using the method developed by Arns and Wiedenbeck  $^{19}$ ), as shown in fig. 6, it is seen that the two solutions of Q, the quadrupole content of the 82 keV gamma ray, are:

$$Q_1 = 0.024 \pm 0.001, \quad Q_2 = 0.924 \pm 0.002.$$

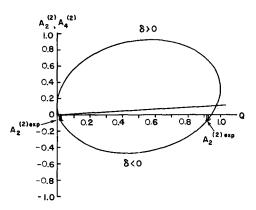


Fig. 6.  $A_2^{(2)}$  and  $A_4^{(2)}$  versus Q, the quadrupole content, for the transition  $\frac{5}{2} \rightarrow \frac{7}{2}$ .

The magnitude of  $A_4^{(2)\text{exp}}$ , although negative, requires one to choose  $Q_1$  as the correct solution. This quadrupole mixture is also in agreement with the predominantly M1 nature of the 82 keV gamma ray deduced from its internal conversion coefficient <sup>9</sup>)  $\alpha = 1.77 \pm 0.05$ . Thus once can conclude that the mixing ratio of the 82 keV gamma ray is  $97.6 \pm 0.1\%$  M1 and  $2.4 \pm 0.1\%$  E2.

Using this mixing ratio and the value <sup>7</sup>)  $6.31\pm0.05$  ns as the half-life of the 82 keV level, calculations analogous to those of Clikeman and Stewart <sup>9</sup>) show that the 82 keV transition is indeed single-particle in character. While its E2 transition rate is enhanced by only a factor ~2 from theoretical single-particle estimates <sup>20</sup>), its M1 rate is retarded by a factor ~ 400. Such a retardation supports the interpretation of the 82 keV level as  $d_{\frac{1}{2}}$ , thus making the 82 keV transition essentially *l*-forbidden  $(d_{\frac{3}{2}} - g_{\frac{7}{2}}; \Delta l = 2)$ .

#### 5.2. THE 302 keV-82 keV CASCADE

The 384 keV level is assigned as the  $d_1$  single-particle state by all recent authors  $^{7-9, 12}$ ). It has been reached by Coulomb excitation 5), and is populated by an electron capture transition from the ground state of Ba<sup>133</sup> (s<sub>1</sub>) with a log ft value <sup>12</sup>) of 7.2. This high value of log ft for an allowed transition suggests l-forbiddenness <sup>8,12</sup>), and supports the assignment of  $d_1$  to the 384 keV level. The 302 keV-82 keV cascade, therefore, has the spin sequence  $\frac{3}{2}^+(D,Q)\frac{5}{2}^+(D,Q)\frac{7}{2}^+$ . Knowing  $A_2 = -0.0238 \pm 0.0060$  and  $A_4 = 0.0061 \pm 0.0089$ , and using  $A_2^{(2)}$  and  $A_4^{(2)}$  for the 82 keV gamma ray obtained from the 356 keV-82 keV correlation,  $A_2^{(1)\text{exp}}$  and  $A_4^{(1)\text{exp}}$  for the 302 keV gamma ray are found to be:

$$A_2^{(1)\text{exp}} = 0.384 \pm 0.098, \quad A_4^{(1)\text{exp}} = -0.8 \pm 1.3.$$

As shown in fig. 7,  $A_2^{(1)\exp}$  gives the following two solutions for Q:

$$Q_1 = 0.003 \pm 0.001, \quad Q_2 = 0.914 \pm 0.029.$$

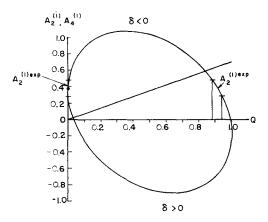


Fig. 7.  $A_2^{(1)}$  and  $A_4^{(1)}$  versus Q for the transition  $\frac{3}{2} \rightarrow \frac{5}{2}$ .

It is obvious that due to the large error in  $A_4^{(1)\rm exp}$ , it cannot serve to help the choice of Q in this case. Neither is the measured internal conversion coefficient for the 302 keV transition known with sufficient accuracy <sup>14</sup>) to facilitate the choice. However, judging from the single-particle interpretation of the 384 and the 82 keV levels, the 302 keV gamma ray should be predominantly M1 in character. It is then concluded that the mixing ratio of the 302 keV gamma ray is 99.7  $\pm 0.1\%$  M1 and  $0.3\pm 0.1\%$  E2.

## 5.3. THE 80 keV-82 keV CASCADE

With  $A_2 = 0.0319 \pm 0.0045$  and  $A_4 = 0.005 \pm 0.009$ , and using  $A_2^{(2)}$  and  $A_4^{(2)}$  for the 82 keV gamma ray from the 356 keV-82 keV correlation results,  $A_2^{(1)\text{exp}}$  and  $A_4^{(1)\text{exp}}$  for the 80 keV gamma ray are calculated to be:

$$A_2^{(1)\text{exp}} = -0.515 \pm 0.077, \quad A_4^{(1)\text{exp}} = -0.7 \pm 1.3.$$

Due to the error in  $A_2^{(1)\text{exp}}$ , this result could fit either a  $\frac{3}{2}(D,Q)\frac{5}{2}(D,Q)\frac{7}{2}$  sequence or a  $\frac{5}{2}(D,Q)\frac{5}{2}(D,Q)\frac{7}{2}$  sequence. However, as will be seen in subsect. 5.4, the 276 keV-162 keV directional correlation measurement has unambiguously determined the spin of the 162 keV level to be  $\frac{5}{2}$ . Therefore, the former sequence is eliminated.

From fig. 8, one obtains the two solutions of Q for the 80 keV gamma ray as:

$$Q_1 = 0.019 \pm 0.019$$
,  $Q_2 = 0.63 \pm 0.09$ .

It will also be shown in subsect. 5.4 that the 162 keV level can no longer be characterized as a single-particle level due to the predominantly E2 character of the 162 keV gamma ray. In view of this result, one would also expect an enhancement of the quadrupole content of the 80 keV gamma ray; and, therefore select  $Q_2$  as the likely solution.

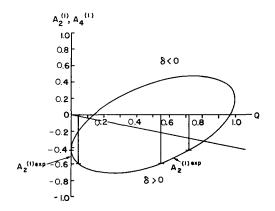


Fig. 8.  $A_2^{(1)}$  and  $A_4^{(1)}$  versus Q for the transition  $\frac{6}{2} \rightarrow \frac{5}{2}$ .

## 5.4. THE 276 keV-162 keV CASCADE

The experimental coefficients for this cascade are:

$$A_2 = -0.328 \pm 0.009$$
,  $A_4 = -0.067 \pm 0.016$ .

Since the spin of the 438 keV level is  $\frac{1}{2}$ , and that of the ground state,  $\frac{7}{2}$ , both  $\frac{3}{2}$  and  $\frac{5}{2}$  are possible values for the intermediate 162 keV level. Therefore, the 276 keV-162 keV cascade could have either the  $\frac{1}{2}(D, Q)\frac{7}{2}(D, Q)\frac{7}{2}$  or the  $\frac{1}{2}(Q)\frac{5}{2}(D, Q)\frac{7}{2}$  sequence.

Assuming  $\frac{1}{2}(D, Q)\frac{3}{2}(Q)\frac{7}{2}$  is the correct sequence, from the table of F coefficients <sup>18</sup>), one obtains:

$$A_2^{(2)} = -0.1428, \quad A_4^{(2)} = 0.$$

The fact that  $A_4^{(2)}$  is zero requires  $A_4$  to be also zero. The experimental  $A_4$  clearly does not satisfy this requirement. Furthermore, the value of  $A_2^{(1)\text{exp}}$  computed from  $A_2$  and  $A_2^{(2)}$  is  $2.297 \pm 0.063$ . Since in the  $A_2^{(1)}$  versus Q graph of the transition <sup>19</sup>)  $\frac{1}{2} \rightarrow \frac{3}{2}$  the highest point of the ellipse has a value of  $A_2^{(1)} = 1.0$ , the  $A_2^{(1)\text{exp}}$  obtained

above cannot possibly fit this spin sequence. Therefore, one is obliged to discard  $\frac{3}{2}$  as a possible value for the spin of the 162 keV level.

In the  $\frac{1}{2}(Q)\frac{5}{2}(D, Q)\frac{7}{2}$  sequence, the F coefficients give:

$$A_2^{(1)} = -0.5345, \quad A_4^{(1)} = -0.6172.$$

Consequently,

$$A_2^{(2)\text{exp}} = 0.614 \pm 0.017, \quad A_4^{(2)\text{exp}} = 0.108 \pm 0.026.$$

Fig. 9 shows that the two values of Q required by  $A_2^{(2)\exp}$  are:

$$Q_1 = 0.123 \pm 0.009$$
,  $Q_2 = 0.951 \pm 0.006$ .

Since the value of  $A_4^{(2)\text{exp}}$  restricts Q to be greater than 0.695, the mixing ratio of the 162 keV gamma ray is therefore  $95.1 \pm 0.6\%$  E2 and  $4.9 \pm 0.6\%$  M1.

The predominantly E2 character of the 162 keV gamma ray indicates that single-particle interpretation can no longer be applied to the 162 keV level.

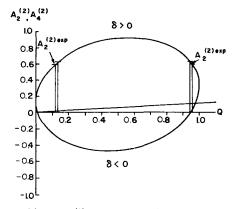


Fig. 9.  $A_2^{(2)}$  and  $A_4^{(2)}$  versus Q for the transition  $\frac{5}{2} \rightarrow \frac{7}{2}$ .

## 6. Summary and Conclusion

The present investigation confirms the decay scheme as shown in fig. 1. The measurements also give support to the predominantly single-particle character of the ground state, 82, 384 and 438 keV levels of  $Cs^{133}$  as  $g_{\tilde{i}}$ ,  $d_{\tilde{i}}$ ,  $d_{\tilde{i}}$ , and  $s_{\frac{1}{2}}$  respectively. The spin of the 162 keV level has been unambiguously determined to be  $\frac{5}{2}$ . Since this level has been reached by Coulomb excitation 5), its parity is therefore +. If an electron capture transition to the 162 keV level did exist, as deduced by Stewart and Lu<sup>8</sup>), then the assignment of  $\frac{5}{2}$  to the 162 keV level would require such a transition to be second-forbidden rather than allowed. Judging from the magnitude of the log ft value  $^{8,14}$ ) (log ft = 9.0) such a requirement does not seem unreasonable. In contrast to the other levels, the character of the 162 keV level cannot be interpreted from the single-particle point of view.

The mixing ratios of the following gamma rays are determined to be: 80 keV:  $(63\pm9)\%$  E2+ $(37\pm9)\%$  M1; 82 keV:  $(97.6\pm0.1)\%$  M1+ $(2.4\pm0.1)\%$  E2; 162 keV:  $(95.1\pm0.6)\%$  E2+ $(4.9\pm0.6)\%$  M1; 276 keV: pure E2; 302 keV:  $(99.7\pm0.1)\%$  M1+ $(0.3\pm0.1)\%$  E2; and 356 keV: pure E2.

Note: During the last stages of this work, an article was published by Münnich, Fricke and Wellner <sup>21</sup>) on the directional correlation measurements in the decay of Ba<sup>133</sup>. The results of the present work, especially with regard to the spin determination of the 162 keV level, agree quite well with the results of these authors.

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