Energy level measurements in ^{158}Gd: An investigation of a possible neutrino balance

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The decay of ^{158}Tb may be a sensitive neutrino balance which could yield the electron neutrino's mass to within a few eV. The determination of the neutrino's mass will require a knowledge of the energy levels in ^{158}Gd and so we present new high precision measurements of the energies of the γ rays from the deexcitation of these levels. They are 780.166±0.007, 897.620±0.055, 944.181±0.009, 962.125±0.009, 977.070±0.170, 1107.635±0.009, and 1187.147±0.007. We also discuss the use of this neutrino balance and we review the current literature regarding the decay of ^{158}Tb.

INTRODUCTION

^{158}Tb has been identified as a possible neutrino balance by the detection of K shell electron capture to the 1187 keV level of ^{158}Gd emitting a neutrino with an energy of only 156±17 eV.[1] The experimental signature for the decay was a coincidence between the K x ray following the electron capture and the γ ray from the deexcitation of the ^{158}Gd (see Fig. 1). If these observations are correct, then the rate of the ^{158}Tb decay will be extremely sensitive to the mass of the neutrino because the total amount of phase space is so restricted. (The only other known electron-capture decay emitting a very low energy neutrino occurs in ^{163}Ho with an energy of approximately 450 eV.[2])

The purpose of this paper is to present new high precision data on the energies of the γ rays emitted after ^{158}Tb decay, and to calculate the low lying energy level structure in ^{158}Gd. These data are essential for the calculation of the neutrino mass as will be discussed in a later section.

EXPERIMENTAL METHOD

We have measured the energy of the 1187.147 keV γ ray to within ±7 eV which is about the maximum precision obtainable by detecting γ rays with a Ge(Li) detector. These measurements were done by comparing the energy of the γ rays emitted by ^{158}Tb (150 years) to an accurately known standard. Care was taken to keep the source a substantial distance away from the detector in order to ensure that the γ rays entered the crystal at the same angle. This improved the energy resolution as the signals had a more uniform risetime than they would have in a close geometry. We also shielded the detector from background radiation with lead and operated the electronics only in their most linear range to enable accurate calibration of the spectra.

R. S. Raghavan provided us with the ^{158}Tb source for this experiment. It was approximately 1 μC of ^{158}Tb and ^{157}Tb in a 50:50 ratio. The ^{157}Tb was not a problem since it only emits low energy x rays and the detector was completely shielded against these by a Ta-Cd-Cu-Al absorber. The detector was a 100 cm^3 Princeton Gamma-Tech Ge(Li) detector. It was enclosed in a Pb house that provided 10 cm of Pb shielding in all directions. The source was placed at various distances from the crystal during different trials but was always greater than 10 cm away.

The detector was calibrated with a ^{56}Co source which was run simultaneously with all of the Tb spectra. Because ^{56}Co has a half-life of only 78.8 days it was made at Caltech by bombarding an ^{56}Fe target with a 9 MeV proton beam from the Kellogg EN tandem Van de Graaff ac-

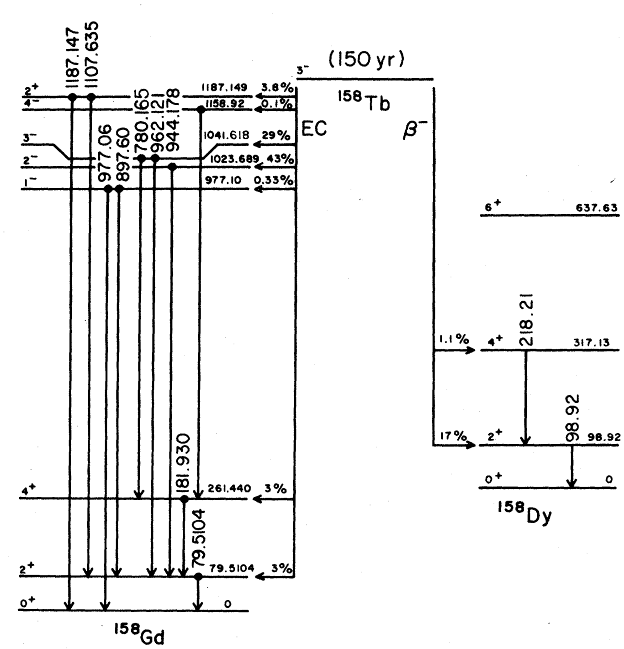

FIG. 1. The ^{158}Tb decay scheme. The γ-ray energies and the energy levels shown here are the recommended values and are the weighted average of our measurements and those of Greenwood *et al.* Table II lists the results separately.

celerator. The source had an initial activity of about 1 μC but also contained very small amounts of ^{57}Co and ^{58}Co. ^{56}Co has several accurately known lines[3] (see Table I) that were used for the calibration of the detector and multichannel analyzer, but it was chosen primarily because of the line at 1175.101$\pm$0.005 keV. This line lies only 12 keV below the 1187 keV Tb line which minimizes the uncertainty in the measurement of this energy. (^{110}Ag^{m} was not used as a calibration standard, for example, because it has a weak line at 1186.7 keV which could not have been separated from the Tb line.)

A thorough examination of the ^{56}Co spectrum revealed that no sum peaks or escape peaks occurred at 1187 keV. The nearest interference occurred at 1191 keV. This small peak was the second escape peak due to the 2212.92 keV Co line. The position and strength of this escape peak was noted and corrected for in all of the Tb spectra. This peak can be seen in Fig. 2 as a small bump at the base of the 1187 keV Tb line. We looked for other sources of background but did not find any significant ones, since long counting trials without sources revealed only a flat background in the region around the 1187 peak.

The relative location of the Co and the Tb sources was important. They both had to be located on the axis of the detector and at the same distance from the detector. Placing the Tb at 10 cm and the Co at 20 cm caused an $\sim$50 eV relative shift between the lines due to each source, presumably due to the different average angle of entrance of the γ rays.

Four trials were run of approximately two weeks duration each. Each trial tested a different amplifier and a different range of channels on a Tracor-Northern model 7200 8K multichannel analyzer. All of the trials were consistent.

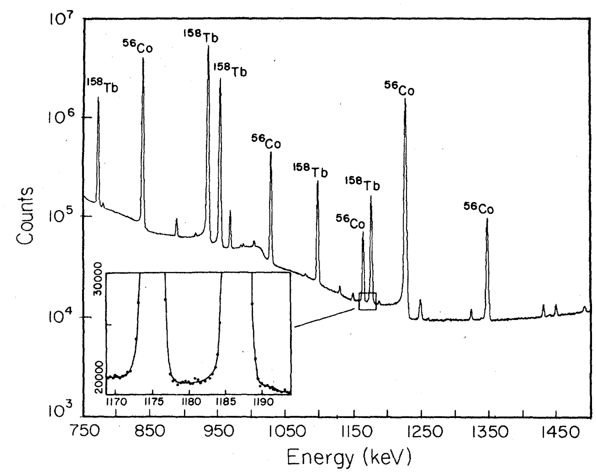

FIG. 2. A typical γ-ray spectrum showing the ^{56}Co calibration lines and the ^{158}Tb lines. The inset shows that a second escape peak from a high energy ^{56}Co line lies close to the ^{158}Tb 1187 peak.

The observed spectra were fit with a 12 parameter function[4,5] of the form

$$y=\frac{\text{Area}}{\sqrt{2\pi}\,\sigma}e^{-(x-\mu)^2/2\sigma^2}\left[1+\text{skew}\left[\frac{x-\mu}{\sigma}\right]^3+\text{skew 1}\left[\frac{x-\mu}{\sigma}\right]^8\right]$$

$$+\frac{\text{Area}}{\sigma}\left[1-\frac{1}{1+[(x-\mu)/\sigma]^2}\right](\text{step}+Ee^{F[(x-\mu)/\sigma]})+A+B(x-\mu)+C(x-\mu)^2+D(x-\mu)^3\ .$$

The first term represents a Gaussian with a well defined mean, area, and width. The second term introduces a small amount of skewing of the Gaussian, the third and fourth terms represent the characteristic step that occurs under a γ line and an exponential tail. Finally, a polynomial background is assumed under the peak. The skewing terms were applied only to channels below the centroid of the peak. Counts falling in channels above the centroid were fit to a pure Gaussian line shape.

Each peak in each spectrum was individually fit using all 12 parameters. The parameters usually showed a smooth variation with energy and a linear or exponential line was fit to these data.[5] Finally, ten of the parameters were fixed to their respective parametrizations and only the position and area of each peak were used as free parameters in the final fits. This procedure produced fits with a χ^2 value of 1 per million counts in the peak for every peak in every spectrum. The energies of the ^{56}Co peaks were then plotted versus the peak positions for each trial. It was assumed that the resulting curves were continuous and the energies of the Tb lines were read from these plots.

TABLE I. ^{158}Tb γ-ray energies and the adopted ^{56}Co calibration energies. The ^{56}Co energies are the weighted mean of the values given by Meyer and Helmer *et al.*

E_γ (keV)	Reference line (keV)	$\lvert\Delta(E_{\text{ref}}-E_\gamma)\rvert$ (keV)
780.166(7)	846.767(5)	66.601(5)
897.620(55)	846.767(5)	50.853(55)
944.181(9)	846.767(5)	97.414(8)
944.181(9)	1037.843(4)	93.662(8)
962.125(9)	846.767(5)	115.358(8)
962.125(9)	1037.843(4)	75.718(8)
977.070(170)	1037.843(4)	60.773(170)
1107.635(9)	1175.101(5)	67.466(8)
1187.147(7)	1175.101(5)	12.046(5)
	1238.285(5)	
	1360.208(6)	

RESULTS

The measured γ-ray energies are listed in Table II. Since the standard deviation of the centroid of a Gaussian is

TABLE II. The observed γ-ray energies can be used to calculate the energy level structure of ^{158}Gd. Here we compare our results to the work of Greenwood *et al.* The recoil of the nucleus has been taken into account and is $E_R = 3.4\times 10^{-9} E_\gamma^2$ (keV).

This work			Greenwood *et al.*		
Initial state energy (keV)	Final state	γ ray energy (keV)	Initial state energy (keV)	Final state	γ ray energy (keV)
			79.5104(23)	0	79.5104(14)
			261.440(7)	79	181.930 (4)
977.121(50)	0	977.070(170)	977.089(25)	0	977.060 (40)
	79	897.620(55)		79	897.590 (45)
1023.694(9)	79	944.181(9)	1023.650(26)	79	944.090 (50)
1041.619(6)	79	962.125(9)	1041.587(26)	79	962.060 (35)
	261	780.166(7)		261	780.140 (35)
1187.151(6)	0	1187.147(7)	1187.097(29)	0	1187.130 (90)
	79	1107.635(9)		79	1107.630 (70)
				261	925.620(70)

$$\sigma_m = \frac{\sigma}{\sqrt{N}}$$

and the standard deviation of the Ge(Li) peaks was less than 0.8 keV then it should have been possible to achieve 1 eV resolution with only 600 000 counts in each peak. In fact, the linearity of the MCA and amplifiers as well as idiosyncrasies in the detector limits the technique to about 5 eV. The important point is that statistical error is not the dominant source of error in these measurements; all trials were run until the peaks of interest had more than 300 000 counts. Some peaks contained 20 000 000 counts. The errors quoted in Table II are therefore standard deviations of the mean of the four measurements added in quadrature to the uncertainty in the energy calibration.

Table II also compares the results of these experiments with the previously accepted energies[6] for the γ rays resulting from the decay of ^{158}Tb. The most important result is that the 1187 line has an energy of 1187.147 ± 0.007 keV as compared to the previous value of 1187.130 ± 0.090 keV. The large uncertainty in the energy of two of the lines listed in Table II is due to an unfortunate overlap between the Tb and Co lines and the fitting procedures could not accurately separate them.

These measurements can probably not be improved upon with a Ge(Li) detection scheme because more accurate standards are not presently available, and presently available detectors are not capable of better stability. These results might be improved upon with a bent-crystal spectrometer.

DISCUSSION

The γ rays resulting from ^{158}Tb decay are indicative of the structure of ^{158}Gd as shown in Fig. 1. The levels at 1187, 1041, and 977 keV all decay by more than one cascading scheme and so this redundancy provides additional data about the energies of these levels. We have calculated the energies of these levels by correcting for the recoil of the nucleus and summing the cascading γ rays. The results are tabulated and compared to the work of Greenwood *et al.* in Table II. The recommended energies would then be the weighted average of our measurements and those of Greenwood *et al.* and these values are used to label the levels in Fig. 1.

We can easily demonstrate that ^{158}Tb would be a sensitive neutrino balance if it decays via K shell electron capture to the 1187 keV level in ^{158}Gd as has been claimed.[1] We will use the notation of Bambynek *et al.*[7] The decay constant for orbital electron capture is

$$\lambda = \frac{G_\beta^2}{4\pi^2} \sum_i Q_i^2 \beta_i^2 B_i n_i C_i \left[1 - M_\nu^2/Q_i^2\right]^{1/2},$$

where G_β is the beta decay coupling constant, M_ν the mass of the electron neutrino, Q_i is the neutrino energy, β_i the Coulomb amplitude of the Dirac radial wave functions, B_i is a factor to account for the imperfect electron wave function overlap and electron exchange, and C_i is the shape factor for beta decay. For closed shells, n equals 1. For partially filled shells, n is equal to the relative occupation number of electrons in the shell. The summation extends over all atomic subshells from which an electron can be captured. The units are defined such that $\hbar = c = m_e = 1$. Bambynek[7] demonstrates that to a very good approximation C_i does not depend on the subshell from which the electron is captured, and that for first forbidden nonunique transitions only the captures from the s and $p_{1/2}$ orbitals are important. Therefore it is useful to define the relative capture probability as the ratio of the K capture decay constant to the total decay constant:

$$P_K = \frac{\lambda_K}{\lambda_{\text{tot}}} = \frac{Q_K^2 \beta_K^2 B_K (1 - M_\nu^2/Q_K^2)^{1/2}}{\sum_i Q_i^2 \beta_i^2 B_i (1 - M_\nu^2/Q_i^2)^{1/2}}.$$

P_K is then independent of the shape factors. Taking the ratio of P_k values to different levels causes all electronic and geometric efficiency factors to cancel out and yields

$$R=\frac{P_K(E1)}{P_K(E2)}=\frac{\text{Yield}_K(E1)}{\text{Yield}_{\text{tot}}(E1)}\frac{\text{Yield}_{\text{tot}}(E2)}{\text{Yield}_K(E2)}$$

$$=\frac{1+\sum_{L,M,N}\frac{Q_i^2(E2)}{Q_K^2(E2)}\frac{\beta_i^2}{\beta_K^2}\frac{B_i}{B_K}}{1+\left[1-\frac{M_\nu^2}{Q_K^2(E1)}\right]^{-1/2}\sum_{L,M,N}\frac{Q_i^2(E1)}{Q_K^2(E1)}\frac{\beta_i^2}{\beta_K^2}\frac{B_i}{B_K}}$$

where the summation extends only over the s and $p_{1/2}$ orbitals in the L,M,N and higher shells and by assumption the neutrino mass is significant only in the decay to the energy level $E1$.

The experimental ratios of yields is therefore subject to direct and simple theoretical interpretation since R depends only on the ratio of the Dirac Coulomb radial wave functions, and the ratio of the electron wave function overlap correction factors. The Q_i are experimentally determined values:

$$Q_i=\Delta M_{\text{g.s.}}-E_x-W_k-E_r\ ,$$

where $\Delta M_{\text{g.s.}}$ is the mass difference between the neutral parent and daughter atoms, E_x is the energy of the level in ^{158}Gd to which the ^{158}Tb has decayed, W_k is the binding energy of the captured electron, and E_r is a rearrangement energy due to imperfect electron wave function overlap in the parent and daughter atoms. E_r is usually very small, of the order of a few eV,[7] and will be neglected in this work.

Raghavan[1] has measured the ratio of K capture yields to the 1187, 944, and 962 levels in ^{158}Gd relative to the 1023 keV sum line. He finds that $R(1187)=(9.0\pm2.0)\times10^{-5}$, and $R(944)-R(962)=(1.09\pm0.15)\times10^{-2}$. The difference of R values was calculated for the 944 and 962 levels in order to cancel out the internal conversion of the cascading 79.5 keV γ ray which could give rise to a false K x-ray coincidence. The implications are interesting. A finite value for $R(1187)$ implies that K capture occurs to the 1187 keV level in Gd. The $R(944)-R(962)$ value implies that the ground state mass splitting between ^{158}Tb and ^{158}Gd is 1242.0±10.0 keV. This is in sharp disagreement with the value for the mass splitting found in the tabulations of Wapstra and Bos.[8] These tables estimate $\Delta M=1216.0\pm1.8$ keV and thus would not allow K capture to the 1187 keV level in Tb.

There are two ways that the discrepancy can be resolved. The data used in the compilation of the mass tables could be wrong or ^{158}Tb has a very complex structure whereby a previously unobserved metastable state at about 20 keV excitation is responsible for the K capture decay to the 1187 keV level in Gd. Burke[9] has checked some of the most important reaction Q values used by Wapstra and Bos to calculate the ^{158}Gd-^{158}Tb mass difference and he finds no significant errors in the data. He has also reexamined the low lying level structure of ^{158}Tb and found that the $K^\pi=4^+$ rotational band head had previously been unobserved and that it occurs at 55.0 keV.[10] This result requires that all of the higher lying levels be shifted up by this amount but he did not find any other low lying levels. His conclusion is that ^{158}Tb is probably not useful as a neutrino balance.

Further investigation, though, reveals that there are data that imply that ^{158}Tb could K capture to the 1187 keV level in Gd. These data are derived from end point measurements of the beta spectrum for ^{158}Tb decaying to ^{158}Dy (see Fig. 1). The end point for the decay to the 2^+ level was measured by F. Schima *et al.*[11] and by Naumann *et al.*[12] some time ago. The end point has been measured again by Altzitzoglou *et al.*[13] with the purpose of reinvestigating Raghavan's result. All of these measurements are summarized in Table III. The weighted average of the end point energies is 851.6±4 keV. Combining this result with the most recent estimate of the ^{158}Gd-^{158}Dy mass difference, 283.0±3.0 keV,[14] yields a ^{158}Gd-^{158}Tb mass splitting of 1233.5±5 keV. This result is close enough to the minimum mass difference, 1237.5 keV, required for K capture to the 1187 keV level in ^{158}Gd to warrant further study.

CONCLUSION

We have measured the energy of the γ rays emitted by ^{158}Tb and we have used these results to calculate the energy of the levels in the daughter, ^{158}Gd. The 1187 keV level in ^{158}Gd may be the final state of an electron-capture decay whose rate is sensitive to the neutrino mass and so we have measured the energy of this level very carefully. It is 1187.151±0.006 keV. This measurement was limited by the accuracy of the γ ray calibration standards available near 1 MeV.

We have also presented a discussion of how ^{158}Tb can be used as a neutrino balance. Because the Tb decay is tagged by both a γ ray and an x ray, we believe that our technique will be easier to implement than the internal bremsstrahlung measurements proposed by De Rujula.[15] Our technique relies on an accurate determination of the transition energy and of the relative K capture rate. We now know one of the quantities required to calculate the transition energy. The only remaining quantity that is not known with sufficient precision is the ground state mass splitting between ^{158}Gd and ^{158}Tb and we believe that this can be measured with the technique of ion cyclotron resonance.[16] We are currently measuring the relative K-capture rate with an apparatus that should be able to determine R to within a few percent or else K shell electron capture to the 1187 keV level in ^{158}Gd does not occur at the reported rate.

TABLE III. The end point energies of the ^{158}Tb to ^{158}Dy β decay as measured by different groups.

Energy (keV)	Error (keV)	Reference
853	5	13
845	10	12
857	27	11
852	8	11

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