Energy of the 2.3-MeV γ ray from the first excited state of ^{14}N

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Using an unusual technique for precisely locating γ ray peaks in a spectrum from a Ge semiconductor detector, the energy of the 2.31-MeV γ ray which follows ^{14}O positron decay has been measured on a scale derived from the known energies of ^{56}Co lines, as have the energies of the 2.18- and 2.32-MeV γ rays which follow the decay of ^{90}Nb. Values of 2312.603(10), 2186.253(10), and 2318.973(10) keV, respectively, were determined. When taken together with updated, previously accepted values, these lead to recommended energies of 2312.590(10), 2186.244(7), and 2318.962(6) keV.

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I. INTRODUCTION

As part of an ongoing program which is measuring the Q values of superallowed positron decays with a precision approaching 10 ppm, we are studying the narrow 1.75-MeV resonance in the $^{13}\mathrm{C}(p,\gamma)^{14}\mathrm{N}$ reaction as a test of our methods. By measuring the proton energy at resonance and also the resulting excitation energy in ^{14}N, we are able to extract a value for the ($^{13}\mathrm{C}+{}^{1}\mathrm{H}-{}^{14}\mathrm{N}$) atomic mass difference and compare it with the accepted value, which is known to a precison of 1 eV from the mass tables. The resonant state in ^{14}N, whose energy we must determine, is at 9.1 MeV, and one of its decay paths to the ground state is via successive γ rays whose energies of 2.7, 2.5, 1.6, and 2.3 MeV are determined relative to the known energies of the γ rays which follow the decay of 77d ^{56}Co.

In the experimental setup, a tightly collimated beam of protons bombards a thin ^{13}C foil mounted on a gold substrate and the γ rays emerge and are detected in a cylindrical, cooled Ge detector. A particular concern is that, since the excited ^{14}N nuclei may be recoiling when they emit the γ rays, the γ energies may be Doppler shifted when seen by the detector. If the axis of the detector is perpendicular to the proton beam direction and looking at the point of impact on the ^{13}C target, then the γ-ray peaks are merely broadened in the spectrum produced by the detector, but if this is not the case, they are Doppler shifted as well. Consequently, it is extremely important that the detector be aligned sufficiently well. For example, in the case being considered, the maximum Doppler shift is 76 ppm/deg.

In Ref. [1] we discussed the difficulties of aligning a Ge detector using surveying techniques, and proposed a method which involved observing the 6- to 7-MeV γ rays emitted from the $^{19}\mathrm{F}(p,\alpha\gamma)^{16}\mathrm{O}$ reaction. Initially the detector is lined up as well as possible using a theodolite, and then, by considering the variation in the asymmetry of the shape of the observed γ-ray peaks as a function of the angle of rotation of the detector about the beam impact point on the target, the perpendicular position can be found. In addition, if the energy of the γ ray is known to high enough precision, then its measurement as a function of detector angle can also be used for alignment.

In the $^{13}\mathrm{C}(p,\gamma)^{14}\mathrm{N}$ case discussed previously, there are similar difficulties with alignment. Studies have shown that the four γ rays of interest are emitted when the ^{14}N nuclei are still recoiling. However, the proper energy of the 2.3-MeV γ ray has been measured with high accuracy by Warburton and Alburger, Ref. [2]. Thus, in the present case, by measuring this γ-ray's apparent energy as a function of the angle of the detector as this is rotated about the beam-target intersection point, one may place the detector at the correct position.

In Ref. [2], the authors adopted the following path to determining the ^{14}N 2.3-MeV energy. First they measured the energy difference $^{90}\mathrm{Nb}(2319)-{}^{14}\mathrm{N}(2313)$. They then found $^{90}\mathrm{Nb}(2319)$ from $^{90}\mathrm{Nb}(2186)+{}^{90}\mathrm{Nb}(133)$, $^{90}\mathrm{Nb}(2186)$ from $^{90}\mathrm{Nb}(2186)-{}^{144}\mathrm{Ce}(2186)$, and $^{90}\mathrm{Nb}(133)$ from $^{57}\mathrm{Co}(136)-{}^{90}\mathrm{Nb}(133)$. So they measured three energy differences, and their final result for $^{14}\mathrm{N}(2313)$ rested on accepted calibration values for $^{144}\mathrm{Ce}(2186)$ and $^{57}\mathrm{Co}(136)$.

Since for the present study of $^{13}\mathrm{C}(p,\gamma)^{14}\mathrm{N}$, the adopted value for $^{14}\mathrm{N}(2313)$ is critical to the establishment of the 9.1-MeV excitation energy in ^{14}N, we thought it worthwhile to remeasure it, based on different γ-calibration lines, those from ^{56}Co, and in addition using a different technique for assigning fiducial positions to γ-ray peaks in spectra from Ge detectors.

II. METHOD

A. Peak fitting in Ge spectra

In Ref. [3], Debertin and Helmer discuss at length many of the algorithms which have been used to fit peak shapes in γ-ray spectra obtained with Ge detectors. The present work sets stringent criteria for such fits. Not only are we aiming to establish the position of a 2.3-MeV peak reliably and repeatably with a precision of a few electron volts, but we must do that for the individual components of the 2313–2319 keV doublet in a situation where the full width at half maximum of a single peak is roughly 3.2 keV. In this case the ability of the chosen algorithm to fit the tails of a peak is obviously particularly relevant. In addition, although it is not involved in these immediate studies, we sought another feature. In the ^{14}N(9.1-MeV) measurements, the energies of the Doppler broadened peaks from the deexcitation γ rays are determined relative to the nonbroadened peaks from calibration sources.

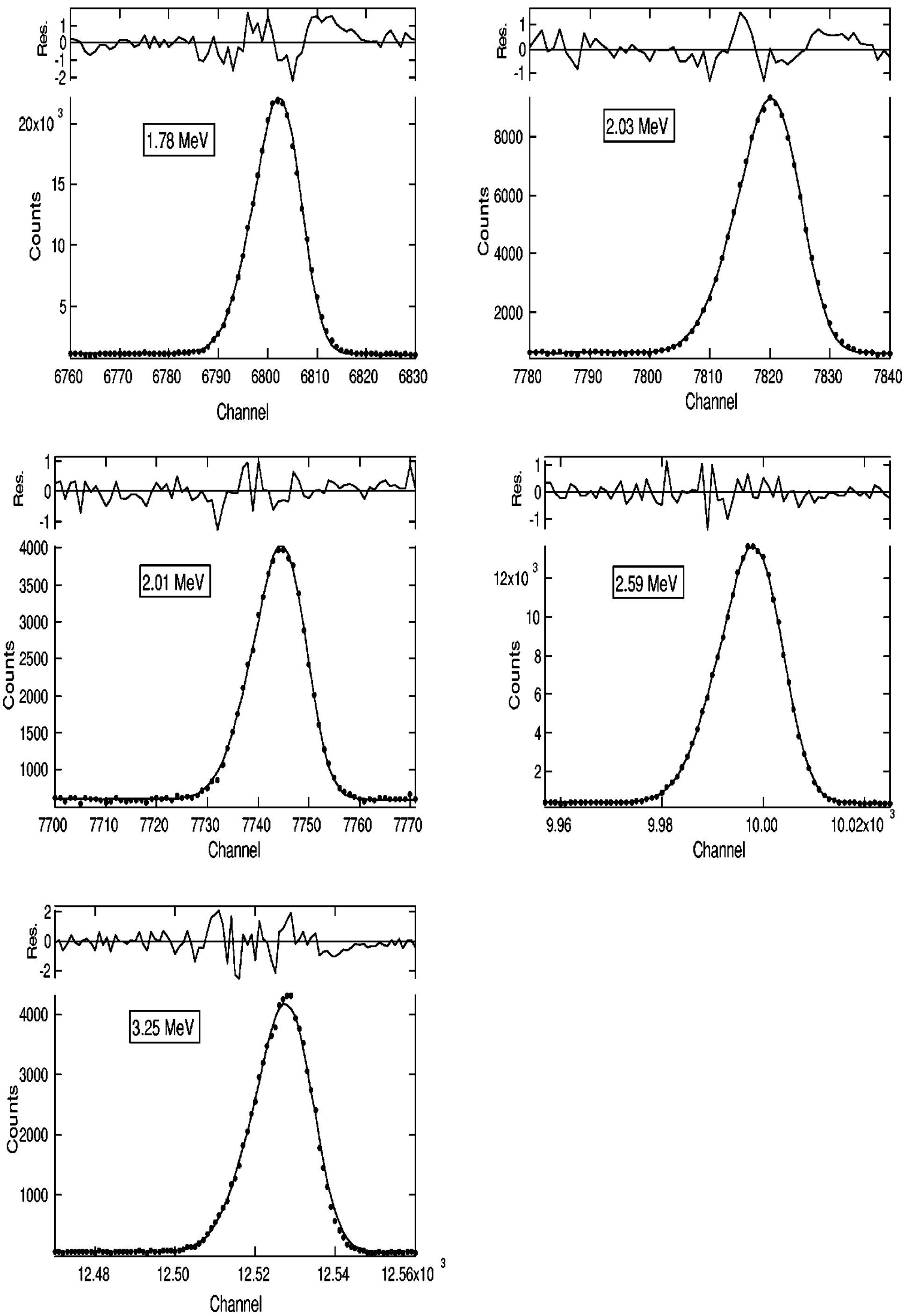

FIG. 1. Fits to the shapes of the ^{56}Co lines at 1.77, 2.01, 2.03, 2.59, and 3.25 MeV, using the shape of the 2.59-MeV line, as described in the text. The residuals are the difference between the data and the fit, divided by the square root of the data.

For this procedure to be valid, the fitting algorithm should find identical positions for a source peak and for the same peak when it is convolved with an appropriate Doppler broadening function.

All the algorithms described in Ref. [3] failed the second test mentioned previously, including the one incorporated in the SAMPO analysis program cited in Ref. [2]. This provided the impetus to develop a new method, which then might, in addition, represent unbroadened peaks in our energy range of interest even more faithfully than earlier methods, and thereby address the problems inherent in doublet fitting discussed previously.

The method adopted is phenomenological and can be illustrated as follows, using the lines from a ^{56}Co source. From the spectrum whose peaks are to be located, choose an isolated, intense, full-energy peak in the energy region of interest, which stands on a constant background. In the present case the 2.59-MeV line is a good choice. If the statistics are only marginally adequate, a low level of smoothing can be applied. Then fit all other lines in terms of the shape of this one using four adjustable parameters: the position, a multiplying factor for the amplitude, a multiplying factor for the width, and a constant background. (Occasionally a sloping background may be needed.) In Fig. 1 the fits

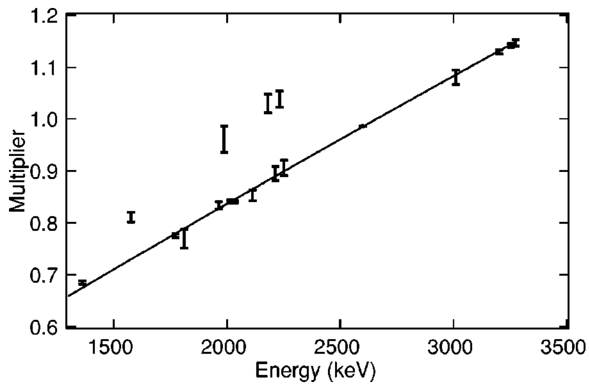

FIG. 2. The width multiplier for all the lines of ^{56}Co from 1.36 to 3.27 MeV, and the fit to this in terms of a parabolic function of the line energy. The four points above the line are from second escape peaks.

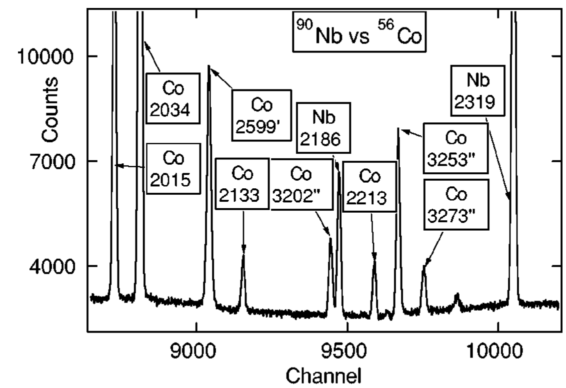

FIG. 3. Part of the energy spectrum of a mixed source of ^{90}Nb and ^{56}Co. The energies in keV volts are indicated, and the $'$ and $''$ designations denote single and double escape peaks, respectively.

to the ^{56}Co peaks at 1.77, 2.01, 2.03, 2.59, and 3.25 MeV using the smoothed 2.59-MeV peak are shown. This is a wider energy range than is required for the present work, but it can be seen that there are very acceptable fits over the whole range.

In this analysis, it would be reassuring if the width multiplier were found to be a smooth and monotonic function of the peak energy. In Fig. 2 this multiplier is shown for all the stronger peaks from 1.36 to 3.27 MeV, from a ^{56}Co γ source. The dependence of the multiplier on energy is seen to be as required, actually nearly linear, and may be represented straightforwardly in terms of a parabola.This has the extra convenience that, if a weak peak is to be fitted, its width multiplier may be fixed at a value derived from the parametrization. The four points in Fig. 2 which do not follow the pattern of the others are from double escape peaks. Interestingly, not only are they as well fitted by the same shape as the others, but their width multipliers are a constant factor, 1.168(4), above the curve for the full energy peaks.

B. γ-ray energy measurements

1. The 2186- and 2319-keV lines from ^{90}Nb

Sources of ^{90}Nb were prepared using the ^{90}Zr$(p,n)^{90}$Nb reaction by bombarding foils of natural zirconium metal with 7.5-MeV protons. Sources of ^{56}Co were prepared using the ^{56}Fe$(p,n)^{56}$Co reaction by bombarding foils of natural iron metal with 6.5-MeV protons. In both cases, more than enough activity could be produced by 10 μA h of protons. The combined radiation was detected in a 17% hyperpure Ge detector, and spectra were taken in 16 384 channels using an ORTEC 572 amplifier and a ND579 spectroscopy ADC. Previous experience has shown that, in intercomparisons of this kind, it is important that the two sources be as nearly as possible at the same point in space, and in the present case they were roughly circular, 5 mm in diameter and on the axis of the detector, 15 cm away from the front face, with roughly 2 cm of lead shielding to reduce the proportion of low energy activity.

The half-life of ^{56}Co is 77 days whereas that of ^{90}Nb is 14 h. Consequently, if a particular spectrum is accumulated over a significant fraction of 14 h, and if there is any dependence of the overall gain of the system on count rate, which in practice is hard to avoid completely, then the energy calibration afforded by the ^{56}Co peaks will not be true for those from ^{90}Nb. To combat this effect, the irradiated zirconium metal was reduced to small grains, and every few hours the material comprising the ^{90}Nb source was augmented to keep the total count rate at a constant level.

Five ^{56}Co– ^{90}Nb intercomparisons were made, with differing source-detector distances and amounts of lead shielding, and with differing amplifier gains to sample different channel regions of the ADC conversion. Part of an initial, exploratory spectrum is shown in Fig. 3, where it can be seen that the portion in which the 2186- and 2319-keV peaks are

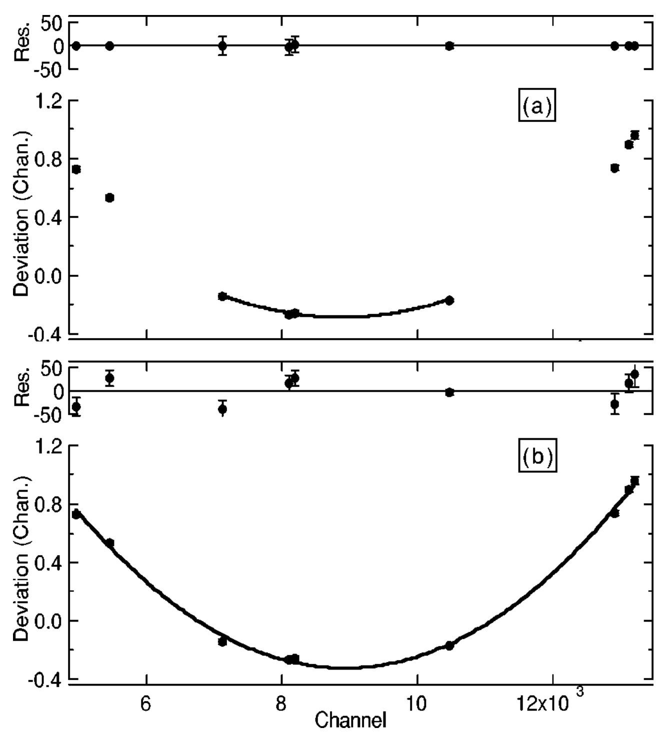

FIG. 4. Short and long calibrations of a γ-ray energy spectrum using the lines from ^{56}Co, as described in the text. The residuals are in millichannels.

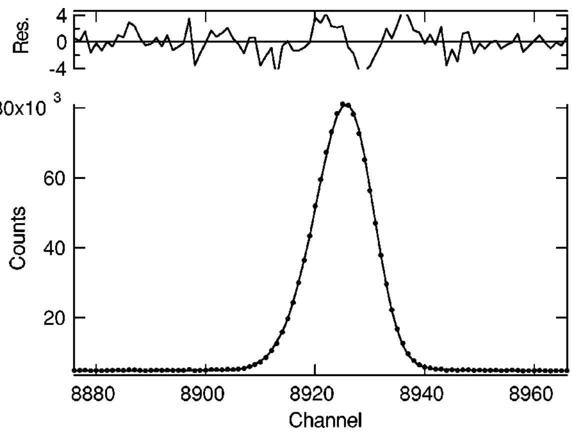

FIG. 5. The fit to the ^{90}Nb 2.319-MeV line using the shape of the ^{56}Co 2.59-MeV line. The residuals are the difference between the data and the fit, divided by the square root of the data.

found is rather crowded, and that in particular the 2186-keV peak is not completely separated from the 3202-keV second escape peak at 2180 keV.

The ^{56}Co calibration peaks chosen were those at 1360.196, 1771.327, 2015.176, 2034.752, 2598.438, 3201.930, 3253.402, and 3272.978 keV, where the values are those cited by Helmer and Van der Leun in Ref. [4]. Each peak was fitted using the 2598-keV peak as a template, in the manner described previously, and a straight line fit was performed to the energy versus extracted peak position dependence. The departure, in fractional channels, of the actual peak position from this straight line was then plotted as a function of channel number, and fitted to a parabola. A typical representation of this parametrization of the system nonlinearity and of the energy calibration is shown in Fig. 4 where the gain of the system is close to 4 chan/keV. Also shown in Fig. 4 is a similar calibration using a restricted energy range from 1771 to 2598 keV. In practice, energies were always obtained using both energy ranges, as a check on consistency. Values rarely differed by more than a few electron volts.

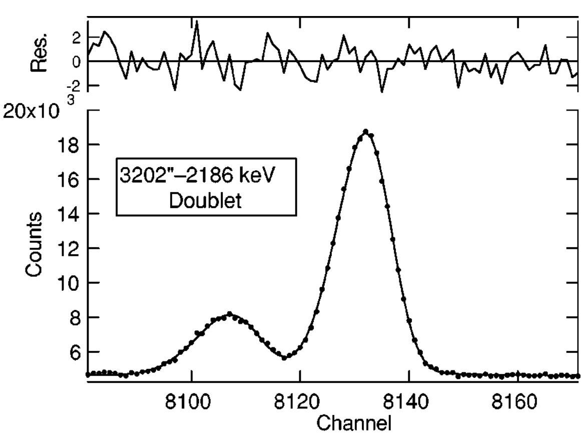

FIG. 6. The fit to the ^{56}Co(3.202″)$-^{90}$Nb(2.186) doublet using the shape of the ^{56}Co 2.59-MeV line. The residuals are the difference between the data and the fit, divided by the square root of the data.

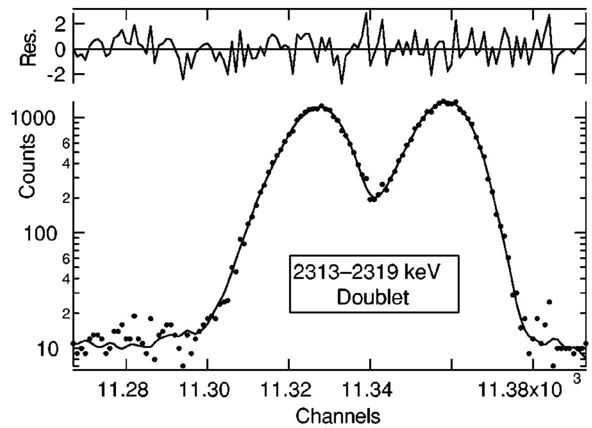

FIG. 7. The fit to the ^{14}O(2.313)$-^{90}$Nb(2.319) doublet using the shape of the ^{56}Co 2.59-MeV line. The residuals are the difference between the data and the fit, divided by the square root of the data.

To extract the positions of the 2186- and 2319-keV peaks, the latter was fitted in the normal way to the standard shape, and the result of this may be seen in Fig. 5. For the former, the 3202–2186-keV doublet was fitted to the sum of two peaks, in which the width of the lower energy component was assigned a multiplier of 1.168 of that of the higher component, as described previously. The result of this procedure for one of the spectra is shown in Fig. 6, and is quite satisfactory, giving an uncertainty in the extracted peak position of 0.05 channels, with a gain of approximately 4 channels/keV.

As a check on most aspects of the overall procedure, the energy of the weak 2213-keV line from ^{56}Co, which sits between the unknown ^{90}Nb 2186- and 2319-keV lines, was determined using the same spectra. The mean of five results was 2212.907(7) keV, which compares well with the value from Ref. [4] of 2212.898(3) keV.

2. The 2313–2319-keV energy difference

The 2313-keV γ ray from ^{14}N was generated using the ^{14}N$(p,n)^{14}$O(β^+) reaction using a tightly focused beam of 7.3-MeV protons. The target was a solid film of elemental nitrogen, kept in vacuum at a temperature of 8 K using a

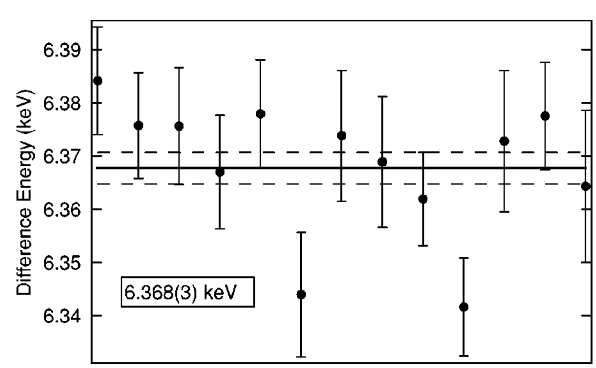

FIG. 8. The results of one analysis of the 13 measurements of the energy difference of the ^{14}O(2.313)$-^{90}$Nb(2.319) doublet, using the shape of the ^{90}Nb 2.19-MeV line.

TABLE I. The energies of the ^{90}Nb (2186-keV), ^{90}Nb (2319-keV), and ^{14}N (2313-keV) lines, and of the [^{90}Nb (2319-keV) − ^{14}N (2313-keV)] difference. All quantities are in keV.

γ transition	This work	Ref. [2]	Ref. [2] updated	Recommended
2186	2186.253(10)	2186.254(10)	2186.237(9)	2186.244(7)
2319	2318.973(10)	2318.968(10)	2318.950(9)	2318.962(6)
2319-2313	6.370(3)	6.375(4)	6.375(4)	6.372(3)
2313	2312.603(10)	2312.593(11)	2312.575(10)	2312.590(10)

Gifford-McMahon two-stage refrigerator, and the beam was regularly pulsed on and off, with the γ rays being detected in the beam off period. The detector was the same one as for the earlier measurements, with its axis on the beam axis and its front face some 12 cm from the target, with 2 cm of lead between to cut down the intensity of the annihilation radiation. The ^{90}Nb calibration source was also on the detector-beam axis, 11 cm from the detector, and once again its strength was adjusted by adding grains of irradiated zirconium metal. The half-life of ^{14}O is 71 s, and so, to avoid the problems with varying count rate alluded to above, the beam on-beam off time periods were set at 10 s, and the system allowed to run for 10 min to come to equilibrium before data taking started.

In all, 13 ^{14}O− ^{90}Nb intercomparisons were made, varying the gain of the electronic system, the source-detector distance, and the relative strengths of the two sources. A typical 2313–2319-keV doublet is shown in Fig. 7, where the fit has been made using the above-described method, with the same, variable width multiplier for both component peaks, and the basic peak shape was from the isolated 2186-keV line. The separation of the doublet, in channels, was converted to keV volts using the positions of the 2186- and 2319-keV peaks. As a check on the robustness of the method with respect to the basic peak shape, the analysis was repeated using the isolated 2319-keV peak from a ^{90}Nb spectrum taken under identical sets of circumstances shortly after each run.

III. RESULTS

The five analyses leading to the energies of the ^{90}Nb lines gave mean results of 2186.253(10) and 2318.973(10) keV, respectively. Both sets of results were internally self-consistent, and the contribution to the ascribed errors of 10 eV from the (very small) errors on the 1772-, 2015-, 2034-, and 2598-keV ^{56}Co calibration lines, which are the ones which principally affect the relevant portion of the energy calibration, are negligible. For each spectrum, the results using the ''short'' calibration were taken, but if the ''long'' calibration results differed, the difference was incorporated in the assigned error.

The 13 2313–2319-keV doublet spectra, in their two analyses, gave mean energy splittings of 6.368(3) keV, with a chi square of 18, and 6.371 keV, with a chi square of 15, of which the former is shown in Fig. 8. A value of 6.370(3) keV was adopted. There was no additonal error introduced from the channel to keV conversions.

To compare the present results with those from Ref. [2], the latter must first be updated in the light of the currently accepted values for the ^{144}Ce and ^{57}Co energies used by the authors as calibrations. The ^{144}Ce line which they took as 2185.662(7) keV is now given in Ref. [4] as 2185.645(5) keV, and this, taken with their measured 2186-keV ^{90}Nb− ^{144}Ce energy difference of 0.592(7) keV, gives the ^{90}Nb line at 2186.237(9) keV. For the energy difference in ^{90}Nb, 2319–2186 keV, they used the ^{90}Nb line at 133 keV, which they determined to be 3.757(2) keV below the ^{57}Co line at 136.4743(5) keV. Since the accepted value for the ^{57}Co energy is now 136.4736(3) keV, Ref. [4], the ^{90}Nb line becomes 132.716(2), and the sum which gives the ^{90}Nb 2319-keV energy becomes 2318.950(9) keV, after correction for recoil effects. Finally, combining this with the 2313–2319-keV doublet separation from Ref. [2], leads to a ^{14}N energy of 2312.575(10) keV, Ref. [5].

The directly comparable ^{14}N energy derived completely independently from the present experimental work is 2318.973(10)–6.370(3) keV, i.e., 2312.603(10) keV.

Perhaps the most justifiable approach is to combine carefully the individual energies from both sets of results. First, the 2186-keV energy is the mean of 2186.237(9) and 2186.253(10) keV, giving 2186.244(7) keV (where, as throughout, more digits have been retained in the calculation than are shown). This may be combined with the energy of the ^{90}Nb 133-keV line from Ref. [2] to give 2318.957(7) keV for the uppermost line. Combining this with the independently determined value from the present work, 2318.973(10) keV, gives a mean of 2318.962(6) keV. Then, the doublet separation is the mean of 6.375(4) and 6.370(3) keV, giving 6.372(3) keV, and the ''best'' value for the ^{14}N energy becomes 2318.962(6)-6.372(3), i.e., 2312.590(6) keV.

These details are summarized in Table I, in which we finally recommend the value of 2312.590(10) keV for the energy of the γ ray emitted by the first excited state of ^{14}N, where we prefer to retain the larger error in recognition of the spread in the values derived via different routes. We also recommend 2186.244(7) and 2318.962(6) keV for the energies of the two ^{90}Nb lines.

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