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GAMMA-RAY ENERGIES MEASURED WITH Ge(Li) SPECTROMETERS

by

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1. Introduction

In this talk, I would like to discuss the capability of spectrometers using lithium-drifted germanium detectors for the precise determination of gamma-ray energies. Of special interest here is the consideration of the problems, and the precision attainable, for gamma rays up to 3 MeV. A few years ago, several people at the NRTS got involved in measuring the energies of some gamma-ray transitions both from radioactive decay and the (n, γ) reaction. These energies typically had uncertainties of 70 eV below 1 MeV up to 0.5 keV above 4 MeV. Some of these energies were determined by adding the energies of the cascade gamma rays. Also in many cases the gamma-ray energies were measured by comparing a double-escape (DE) peak with nearby full-energy (FE) peaks. In the course of these experiments, it became clear that any further improvement in these energy measurements would have to be preceded by a systematic review and analysis of the assumptions involved.

MASTER

In this paper, I will give a summary of a study to measure and compare gamma-ray energies over the range of 50 to 3500 keV. This includes a review of analysis procedures for data from Ge(Li) spectrometers as well as some of our results both published and preliminary. For convenience, I have broken the rest of this paper into three sections. The first is a discussion of the energy scales that are commonly used and their compatibility. The second is

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a discussion of techniques; both those generally used and the specific ones we have used. This section includes some examples of our results up to 3500 keV. Steps that may lead to a significant reduction in the uncertainties in the gamma-ray energies are noted. The last section includes comments on the problems resulting from the field effect and those involved in the use of DE peaks.

2. Energy Scales

In gamma-ray spectroscopy, energies have been commonly measured on two energy scales. The first scale has used the energies, or wavelengths, of the atomic x-rays for calibration and reference. This "x-unit" scale was used initially to make the best use of the high precision that was obtained with crystal diffraction spectrometers. Currently these measurements are often based on the wavelength scale suggested by Bearden (1967). In this work, the previous intercomparisons of the x-ray wavelengths were re-analyzed in order to obtain the most consistent and accurate set of energies possible. For this energy scale, Bearden chose to use the W $K\alpha_1$ line as the reference; he quoted a value of 59.318 24 keV for this line. From the adjustment of the fundamental constants by Taylor et al. (1969), we obtain an adjusted W $K\alpha_1$ x-ray energy of 59.319 18 keV (± 5.9 ppm). It might be noted that one problem with the use of x-ray lines for calibration is their intrinsic width.

The second energy scale that is in common use is based on the energy corresponding to the rest mass of the electron, $m_0 c^2$. The electron mass is derived in the adjustments of the fundamental constants, and in the latest adjustment [Taylor et al. (1969)] its value is quoted as 511.0041 keV (± 3.3 ppm). Although this unit of energy is, in principle, directly available for calibration in any process involving the annihilation of a positron-electron pair, this annihilation line is generally not satisfactory for energy measurements.

Since most positrons annihilate in matter with bound electrons, the requirement that both momentum and energy be conserved in the reaction leads to a line that is a few keV wide. Also, the transfer of energy to the bound electron and to the atom, shifts the observed photon energy to a somewhat lower and uncertain value. One method of overcoming this problem is to use the narrow (but very weak) component of the annihilation line that results from the annihilation of positronium. Such measurements must be carried out with a very high resolution spectrometer and with the annihilation in a material that maximizes the positronium production. The only measurement of this type is that of Murray et al. (1965) in which the energy of the 411-keV gamma ray from the decay of ^{198}Au was measured with respect to this narrow component by means of external-conversion-electron spectroscopy. Since all subsequent measurements on this energy scale have been made with respect to the energy of the 411-keV line, the effective uncertainty in this scale is the total error of the 411 energy, which we take to be 19 ppm.

The question that arises next is whether these energy scales are in agreement. The results of six comparisons of these scales have been summarized in Table I. These results have been corrected to correspond to the 1969 values of the fundamental constants [Taylor et al. (1969)]. Note that three values are direct comparisons of the annihilation and $W\text{ K}\alpha_1$ wavelengths while the others involve the 411-keV line from ^{198}Au . Although the most precise of the older scale comparisons indicates a discrepancy of 56 ± 14 ppm, Van Assche et al. (1971) have shown that in this experiment, the 511-keV line is actually a complex of several lines. Excluding this value, all of the others indicate that the energy scales are consistent, at least within the accuracy of a Ge(Li) spectrometer.

3. Gamma-Ray Energy Measurements

A. Philosophical Divergence

Before describing our procedure, it would be worthwhile to note that there are different approaches taken by compilers and measurers of gamma-ray calibration energies. In 1968, Marion had available only the first two of the above comparisons of these energy scales. He, therefore, concluded that the two scales were inconsistent and chose to recommend and compile only the energies based on the $m_0 c^2$ or 411-keV value.

Another procedure was followed recently by Kern (1972) in compiling a set of gamma-ray energies. He concluded the two energy scales are consistent to 10 ppm and accepted values from both scales. However, as primary standards above 300 keV, he chose to use only the energies measured on the Chalk River electron spectrometer. Although this method certainly provides the maximum consistency, it may not provide the most accurate values. In contrast, we have chosen to include all previous precise measurements. This does, of course, make the standards less internally consistent, but we hope they will be more accurate.

B. General Methods

Our gamma-ray energy measurements have been made on several different Ge(Li) spectrometers involving planar, true coaxial, and five-sided coaxial detectors with volumes from 2 to 50 cm³. All of these detectors have preamplifiers with field-effect transistors in the first stage; some have cooled and others have room-temperature first stages. Each system uses a high-quality linear amplifier and a 4096-channel analog-to-digital converter. All systems have good energy resolution; that is, a full-width-at-half-maximum of ≤ 1.2 keV at 100 keV and < 2 keV at 1 MeV. This good resolution is of value primarily so that we can measure the energy differences between lines that are just a few keV apart. In all cases, the deviation from linearity

was less than ± 2 channels from channel 200 to 4000.

In figure 1 is shown a typical spectrum for our energy measurements. Note that the number of calibration lines is large. There are four steps in the analysis of such a spectrum:

1. the determination of the location of the peaks of interest,
2. the correction of the peak positions for the non-linearity of the system,
3. the determination of the energy vs. channel function $E(x)$ from the energies and positions of the calibration peaks, and
4. the computation of the gamma-ray energies and uncertainties; or the calculation of the difference in energy of specific pairs of peaks.

These four operations are carried out as follows:

In order to determine the position (and area) of a peak, it is usually fit with either a simple Gaussian (plus some function to represent the background) or a more complex function. Since we have been primarily interested in the peak positions, we have used a simple Gaussian function. The background is taken as a straight line (generally of zero slope) matched to the average count in the background on the high-energy side of the peak. The computer program used for this analysis includes a routine which automatically locates the peaks.

The precision of the energy measurements is limited in part by the errors in the peak locations. For the spectrum in figure 1, the computed errors in the peak positions of interest are 6 to 50 millichannels with a mean of 18 millichannels. (This is less than 1/100th of the peak width.) Since only 5 to 9 channels were used in these peak fits, the extremely small peak location errors (i.e., < 20 millichannels) may be fortuitous. However, if they are, this fact should show up when the average value is computed for several measurements.

As noted above, the second step in our computation of the gamma-ray energies is the correction of the peak positions for the nonlinearity of the system. For each spectrometer, this correction is obtained from spectra of several sources that have gamma rays whose energies are well known. From the peak positions and energies of two of these lines, a straight line is determined that would represent the energy if the system were linear through these two points. Then for all of the peaks the differences between the energies computed from this line and the actual values, when converted to channels, give the deviation from linearity. By making these measurements with different gamma rays (i.e., different isotopes) and at different amplifier gains, the effects of the errors in the gamma-ray energies are averaged out. An example of such data taken at three gains is shown in figure 2. The nonlinearity correction used is taken from a smooth curve drawn through the experimental points.

The question of the accuracy of the nonlinearity correction is a complex one. Even at the time of the linearity measurements, this curve is probably not defined to better than 50 millichannels. Since this value is larger than the uncertainty in the peak positions, this is of some concern. Also since a smooth curve is used to represent the nonlinearity correction, any local fluctuations in the nonlinearity will not be taken into account.

In our measurements, the following steps have been taken to minimize the effects of errors in the nonlinearity correction. Most of our measurements are of energy differences so that the long-ranged errors have only a very small effect. Second, each energy determination involves measurements taken as several different gains, in order to average out any effects from local fluctuations in the nonlinearity. Also, most of the results involve measurements taken from two to four different spectrometers. The agreement of the

results from the different systems is a verification of the quality of the linearity corrections.

The third step in the determination of the gamma-ray energies is the computation of the energy vs. channel function $E(x)$ which is taken to be either linear ($a + bx$) or quadratic function ($a + b \cdot x + c \cdot x^2$). The coefficients a , b and c are computed from a weighted linear least-squares fit to the energies of the calibration lines. Since both the calibration energies and channels have uncertainties, the most correct procedure would be to do a bi-variate fit; however, we have chosen a less exact but faster method. If the peak positions were exact, the weights for the E vs. x fit would be computed from the calibration line errors as simply $1/\sigma^2(E)$. The error in the peak position is taken into account in an approximate way by converting it to energy units and adding it in quadrature to $\sigma(E)$; that is, the weights are $1/[\sigma^2(e) + b^2\sigma^2(x)]$.

The matrix that is computed and inverted in order to obtain the parameters a , b and c also contains the information on the associated variance and covariance terms. Finally these parameters and associated error terms as well as the peak positions and uncertainties are used to determine the energies and uncertainties of the gamma rays.

C. Specific Methods and Results

We have two gamma-ray energy measurement papers in print. Our first group of energy measurements [Greenwood et al. (1970)] were concerned with the energy region below 400 keV. In this region there have been many precise measurements, both with crystal diffraction and electron spectrometers. Therefore, our major effort was to compare the existing energies to determine their compatibility. We found the energies compatible to the precision that we could measure. Therefore, we have used these values as a basis for work at higher energies.

Our second group of gamma-ray energy measurements [Helmer et al. (1971)] cover the energy range from 400 to 1300 keV. In this region the number of lines available from long-lived radioactive sources whose energies have been reported with an accuracy of 35 ppm or better was not sufficient for the accurate calibration of Ge(Li) spectrometers. Therefore, we determined the energies of a number of new calibration lines in this region.

Since Ge(Li) spectrometer systems are inherently nonlinear, we did not consider it satisfactory to rely on interpolation over a large energy, or channel, range even with the use of a nonlinearity correction. Two techniques were used in order to avoid such interpolations. First, we used cascade-crossover combinations where we were able to determine accurately the energies of two of the three transitions (usually the cascade transitions), so that the third energy could be computed. The second technique involved the measurement of small energy differences (less than 3% of the energy, or 30 keV at 1 MeV). Figure 3 is a portion of a spectrum illustrating the measurement of four energy differences relative to the 884-keV line from ^{192}Ir .

Figure 4 shows a diagram of two chains of energies that have been built up to ~ 1300 keV by the use of these techniques. The cascade-crossover combinations and the energy differences measured are indicated. One chain is based on the transitions from ^{192}Ir with ^{160}Tb and the other chain is based on those of ^{198}Au with ^{182}Ta and ^{59}Fe . Since the ^{192}Ir energies have been determined primarily on the m_0c^2 (i.e., ^{198}Au 411 keV) scale, both of these chains are on this scale. In addition to providing two independent routes up to 1300 keV, these chains provide a method of accurately testing for the accumulation of systematic errors in the energies of the chains. One such test is illustrated in figure 5 where spectral data for a 1177 (^{160}Tb)-1189 (^{182}Ta) energy difference measurement are shown. From comparisons of the energies of the two chains involving three such pairs of peaks, it was determined that

there was a discrepancy in the preliminary energies of 17 ± 4 eV. (See Ref. 8 details of this calculation.) Our final reported energies were adjusted to remove this discrepancy.

For the data in figure 1, the average peak position uncertainty was about 20 millichannels. If this value applies to a pair of 1 MeV peaks at channel 3000, one spectrum gives an uncertainty of ~ 10 eV; and the average of 10 measurements gives ~ 3 eV (or 3 ppm of the gamma-ray energy). This estimate agrees with the results for the two energy differences shown in figure 5, as indicated in Table II. In both cases, the data from the different spectra and the different detectors are completely consistent. That is, the values

of ϵ^2
$$\left[\text{i.e., } \frac{1}{N-1} \sum_{i=1}^N \left(\frac{E_i - \bar{E}}{\sigma_1(E)} \right)^2 \right]$$
 for the 14 and 29 measurements are

both ~ 1.0 (namely, 0.6 and 1.1). It should also be noted that the computed errors in these differences are both ± 2 eV. (This value does not include any systematic error that would be common to all of the detector systems.)

From the energies of the transitions in the two chains up to 1300 keV shown in figure 4, other line energies were determined from small differences and cascade-crossover combinations. This system of energies was then extended to 3500 keV by the same methods. Of the ~ 100 energy differences measured below 1300 keV, most have uncertainties of < 5 eV. And of the ~ 35 differences between 1300 and 3500 keV, most have uncertainties < 10 eV. In computing the total error in a new gamma-ray energy, this measurement uncertainty must be combined with the measurement error in the reference line (of the pair) as well as the uncertainty in the energy scale. This process has resulted in energies for ~ 75 transitions in the 400 - 1300 keV region and for ~ 40 transitions above 1300 keV. Table III gives the preliminary results for two

isotopes which extend above 1300 keV. The isotopes for which we have measured energies in this region include ^{58}Co , ^{82}Br , ^{84}Rb , ^{60}Co , ^{124}Sb , ^{88}Y , ^{144}Ce , ^{24}Na , ^{132}Cs , in addition to those in Table III. The associated errors are 21 - 31 ppm with the dominant contribution from the 19 ppm error in the energy scale. This fact suggests that a significant reduction in these errors would be forthcoming if the energy of the ^{198}Au 411-keV line were improved. However, an even more dramatic improvement could be accomplished if two lines, such as the 411- and 675-keV lines from ^{198}Au , were accurately determined (to say 1 ppm). Then our energy differences could be used to generate a whole set of calibration lines up to 3500 keV, some with uncertainties of ~ 5 ppm. (The work of Deslattes (1971) and Sauder (1971) at the National Bureau of Standards may provide such calibration lines.)

4. The Field Effect

For the past few years, it has been well known [Gunnink et al. (1968), Heath (1969)] that the primary electrons from gamma-ray interactions are accelerated by the electric field in a Ge(Li) detector. This effect can be quite large due to the fact that for all except low-energy gamma rays, the primary electrons are emitted preferentially in the direction of the gamma ray. Typical values of the shift in the apparent peak energy are shown in Table IV for a few energies and for three detector configurations. These measured values are based on the assumption that there is no acceleration if the gamma rays enter a planar detector perpendicular to the electric field. As shown in the table, the observed shift is ~ 100 ppm for these planar and closed-coaxial detectors for FE peaks and about a factor of 10 smaller, and in the opposite direction, for DE peaks.

In our energy measurements we have, so far, used only FE peaks. Even in this case, attention must be paid to the field effect. First, all of the

sources must be at the same angle to the electric field. As a second order effect, this means that all the sources should also be at the same distance from the detector. Table V shows the effect on the apparent peak energy of changing the source-detector distance from 15 cm to 1 cm for two detectors. For the closed-coaxial detector at 2600 keV the shift is ≈ 110 eV and is still ≈ 40 eV at 700 keV. These values are very large compared to the errors in the energy differences quoted earlier. Therefore, for precise measurements, one must either make sources that can be compared at similar (and preferably, large) distances, or one must correct for the variation in the field effect for the geometries used.

For DE peaks, the question is even more complex. As noted in Table IV, the FE and DE peak shifts are different. Therefore, if precise comparisons of the energies of FE and DE peaks are to be made, the radiations must both enter the detector perpendicular to the electric field, or accurate corrections for the field effect must be made. The former method is not possible for closed-coaxial detectors.

The second question that arises with respect to the use of DE peaks for energy measurements is whether the DE-FE peak separation for one transition is exactly $2m_0c^2$. In the pair production process, a certain fraction of the positrons will annihilate with bound electrons. This means that some of the $2m_0c^2$ energy will be left in the detector as the two annihilation photons escape. To the extent that this energy produces ion pairs (or holes), the energy of the DE peak will be shifted to a somewhat higher energy. Therefore, it would seem very desirable to measure the FE-DE peak energy difference. A previous measurement [White et al. (1968)] has been carried out; it allows this difference to be as much as 50 eV less than $2m_0c^2$. We have made one comparison involving three transitions from ^{124}Sb (namely 645, 1045 and 1691 keV.) Measurements

were made of the 1045-keV energy and the difference between the 645-keV FE peak and the 669-keV DE peak (from the 1691-keV transition). The latter measurement was made with the gamma rays entering a planar detector perpendicular to the electric field. From the preliminary results of this experiment, we find that the FE-DE peak difference is less than $2m_0c^2$ by 15 ± 25 eV. If this preliminary result proves to correct, we can use the DE peaks to extend our energy calibration set to higher energies with an uncertainty of ≈ 25 eV/MeV. (This assumes that the field effect has been eliminated.) This would be quite satisfactory for the present, since our m_0c^2 energy scale (via ^{198}Au 411 keV) has a basic uncertainty of 19 eV/MeV.

Therefore, it would seem that with the present energy scales if one accepts the philosophy of this method and the results so far, we can expect to establish a set of calibration energies with uncertainties of ≈ 25 ppm.

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TABLE I

COMPARISON OF THE W $K\alpha_1$ AND ANNIHILATION BASED ENERGY SCALES

Reference	Method of Comparison	Increase in W $K\alpha_1$ Energy Needed to Bring Energy Scales into Agreement (ppm)
Knowles (1962)	$\lambda(m_0 c^2) - \lambda(W K\alpha_1)$	-8 ± 32
Knowles (1963)	$\lambda(m_0 c^2) - \lambda(W K\alpha_1)$	57 ± 14
Reidy & Wiedenbeck (1966)	$\lambda(411) - \lambda(W K\alpha_1)$	-18 ± 42
Greenwood <u>et al.</u> (1970)	$E_{411} - E_{406} (^{183}\text{Ta})$	0 ± 28
Van Assche <u>et al.</u> (1971)	$\lambda(m_0 c^2) - \lambda(W K\alpha_1)$	14 ± 33
Piller <u>et al.</u> (1973)	$\lambda(411) - \lambda(W K\alpha_1)$	2 ± 11

TABLE II

ENERGY DIFFERENCE MEASUREMENTS

Transitions	Detector	Number of Measure-	Energy Difference (eV)	Average Difference Value c^2 (eV)
1189(^{182}Ta) - 1177(^{160}Tb)	P-40 P-60 ND-2	6 6 2	11082 $\pm$ 4 11088 $\pm$ 3 11086 $\pm$ 9	0.6 11085 $\pm$ 2
1221(^{182}Ta)- 1231(^{182}Ta)	P-31 P-40 P-60 ND-2	5 6 16 2	9612 $\pm$ 4 9608 $\pm$ 5 9603 $\pm$ 2 9603 $\pm$ 9	1.1 9606 $\pm$ 2

TABLE III

Gamma-Ray Energies Above 1 MeV from ^{110m}Ag and ^{56}Co

Preliminary Values

<u>Parent Isotope</u>	<u>Method</u>	<u>Average Energy (keV)</u>
^{110m}Ag	706 + 677 SUM	1384.267 $\pm$ 0.029
	763 + 620 SUM	
	937 + 446 SUM	
	818 + 657 SUM	1475.757 $\pm$ 0.034
	818 + 687 SUM	1505.006 $\pm$ 0.032
	884 + 620 SUM	
	818 + 744 SUM	1562.264 $\pm$ 0.033
	884 + 677 SUM	
^{56}Co	1037-1043(^{82}Br) DIFF	1037.216 $\pm$ 0.022
	-1063(^{207}Bi) DIFF	
	1175-1189(^{182}Ta) DIFF	1175.067 $\pm$ 0.026
	-1121(^{182}Ta) DIFF	
	1238-1221(^{182}Ta) DIFF	1238.256 $\pm$ 0.027
	1230(^{182}Ta) DIFF	
	1257(^{182}Ta) DIFF	
	1360-1373(^{182}Ta) DIFF	1360.175 $\pm$ 0.029
	1384(^{110m}Ag) DIFF	
	1771-1836(^{88}Y) DIFF	1771.300 $\pm$ 0.037
	1810-1836(^{88}Y) DIFF	1810.665 $\pm$ 0.039
	1963-1897(^{84}Rb) DIFF	1963.675 $\pm$ 0.040
	-2002(^{125}Sn) DIFF	
	2015-1897(^{84}Rb) DIFF	2015.137 $\pm$ 0.040
	2002(^{125}Sn) DIFF	
	2034-1897(^{84}Rb) DIFF	2034.706 $\pm$ 0.041
	-2185(^{144}Ce) DIFF	
	-2002(^{125}Sn) DIFF	
	2113-2002(^{125}Sn) DIFF	2113.049 $\pm$ 0.045
	2185(^{144}Ce) DIFF	

Table III (continued)

<u>Parent Isotope</u>	<u>Method</u>	<u>Average Energy (keV)</u>
⁵⁶ Co	2212-2185(¹⁴⁴ Ce) DIFF	2212.861 ± 0.049
	1238 + 1360 SUM	2598.400 ± 0.053
	1238 + 1963 SUM	3201.901 ± 0.064
	1238 + 2015 SUM	3253.361 ± 0.065
	1238 + 2034 SUM	3272.927 ± 0.065
	1238 + 2212 SUM	3451.064 ± 0.069
	1037 + 1175 + 1238 SUM	

TABLE IV

Peak Energy Shifts for Various Detector Configurations

Detector		Gamma-Ray Energy (keV)	Peak Shift (eV)		
Type	Voltage		FE Peak	De Peak	Relative FE-DE Peak
Planar	1900	820	32 ± 6		
		1180	76 ± 4		
		1680	114 ± 18	-18 ± 11	132 ± 21
		2180			255 ± 30
		2600	264 ± 16	-23 ± 9	287 ± 18
True Coaxial	2500	2180			25 ± 30
Closed Coaxial	2000	2180			300 ± 40

TABLE V

Peak Energy Shifts for Source Positions of 1 and 15 cm

Detector		Gamma-Ray Energy (keV)	FE Peak Shift (eV)
Type	Voltage		
True Coaxial	2500	2600	-37 ± 28
Closed Coaxial	3300	700	42 ± 11
		1490	58 ± 10
		2600	113 ± 29

Figure Captions

1. Typical energy calibration spectrum for determination of the energies of the four lines from ^{94}Nb and ^{95}Zr . Energies are in keV. The calibration peaks are shaded.
2. Data from three gamma-ray spectra for the determination of the non-linearity correction for a typical Ge(Li) detector system.
3. Portion of a gamma-ray spectrum illustrating the energy difference between four transitions and the 884-keV line of ^{192}Ir .
4. Diagram of gamma-ray transitions used in energy chains up to 1300 keV. The measured energy differences and energy values determined are shown.
5. Portion of a gamma-ray spectrum illustrating the energy difference measurements near 1200 keV including a comparison of the two energy chains [$^{1177}({}^{160}\text{Tb})$ - $^{1189}({}^{182}\text{Ta})$].

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Fig. 1

Fig. 2

Fig. 3

Fig. 3

^{198}Au , ^{182}Ta , ^{59}Fe Set

Known Energies

^{198}Au : 411, 675

^{182}Ta : 67

^{59}Fe : 192

Measured Differences

$$D_1 = 1121 - 1087$$

$$D_2 = 1099 - 1087$$

Computed Energies

$$^{198}\text{Au} : 1087 = 411 + 675$$

$$^{182}\text{Ta} : 1121 = 1087 + D_1$$

$$1189 = 1121 + 67$$

$$^{59}\text{Fe} : 1099 = 1087 + D_2$$

$$1291 = 1099 + 192$$

^{192}Ir , ^{160}Tb Set

Known Energies

^{192}Ir : 295, 308, 316

^{160}Tb : 93, 298

Measured Differences

$$D_3 = 604 - 588$$

$$D_4 = 612 - 588$$

$$D_5 = 884 - 879$$

Computed Energies

$$^{192}\text{Ir} : 604 = 308 + 295$$

$$612 = 316 + 295$$

$$588 = \begin{cases} 604 - D_3 \\ 612 - D_4 \end{cases}$$

$$884 = 588 + 295$$

$$^{160}\text{Tb} : 879 = 884 - D_5$$

$$1177 = 879 + 298$$

$$1271 = 879 + 298 + 93$$

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Fig. 5