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GAMMA-RAY STANDARDS

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Abstract

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This paper reviews gamma-ray standards which are used for the calibration of Ge(Li) detectors. A critical analysis is presented of the available reference standards: W K_{α_1} X-rays, the ^{198}Au 411 keV transition and m_0c^2 (annihilation radiation). It is seen that the presently accepted energies are consistent within about 10 ppm.

γ-rays which have been calibrated with special precision directly against one of the two above-mentioned reference standards are defined as primary standards, only data obtained with iron-free magnetic spectrometers or crystal spectrometers qualify for this designation. A survey of the most precise results is performed. There seems to be a systematic discrepancy between the data of Chalk River and those obtained with crystal spectrometers. Since the former results are of proven consistency over a fairly large energy range, the Chalk River set of primary standards for energies over 280 keV is adopted. The errors quoted by the authors are not always unambiguous, and this subject is discussed in a separate section. Secondary standards have been obtained from the primary standards by several methods, which are presented. Special attention is given to the consistency requirement in setting up series of standard lines. Such consistency should result in a better agreement for (true) energy combinations. A short compilation of recent data on some widely used isotopes is presented.

The problem of high-energy standards (E_γ > 2 MeV) is discussed. In this energy range, ^{56}Co has emerged as the most frequently used radioactive source for both energy and intensity calibrations; a compilation of recent results is given. The general question of intensity calibration is the subject of a special section.

Finally, an outlook on the current developments, which will contribute to important improvements in the accuracy of energy standards, is given.

1. INTRODUCTION

Working with Ge(Li) detectors, it is possible to determine the position of peaks with good statistics with a precision corresponding to a few electron-volts (eV). This accuracy is meaningful only if the non-linearity of the detection chain can be determined with comparable precision. Investigators have proposed a number of γ-ray energy standards. The use of the published values needs some caution because they do not always refer to the same energy scale, and the meaning of the quoted errors is not unambiguous. A discussion on reference standards and on errors is given in sections 2 and 3. Primary standards are surveyed in section 4. Methods of obtaining secondary standards and results for γ-rays of energy < 2 MeV are presented in section 5. A review of standards for higher energies and for intensity calibration is the subject of sections 6 and 7. In the conclusion an outlook is given on the developments expected in the near future.

2. ENERGY SCALES

For both historical and physical reasons, the energies of γ-rays are related to X-ray standards or to the electron rest-mass energy. The accepted values of these energies have fluctuated in the course of time.

2.1. X-ray standards

X-ray wavelengths have been measured for several decades in X-units (xu), a quantity proposed by M. Siegbahn and intended to be 10^{-3} A. The xu was defined as the first grating constant of calcite (d_1 = 3029.04 xu). This definition turned out to be inadequate, because even so-called "perfect" calcites gave variations of the order of 20 ppm [1]. It was then suggested by DuMond [2] and Bergvall et al. [3] that an emission line would yield a much better definition of the length unit. The adopted value for the Mo K_{α_1} = 707.831 xu disagrees, however, [4] with the calcite d_1 = 3029.04 xu definition and also with the accepted Cu K_{α_1} xu value. In view of all these discrepancies, data based on X-ray standards have been simply disregarded by Marion [5] in his compilation of γ-ray standards. By Bearden [6, 7] then a new wavelength standard has been introduced, the so-called $\mathring{A}^*$, defined by the errorless relation

$$\lambda(W\ K_{\alpha_1}) = 0.209\ 0100\ \mathring{A}^* \tag{1}$$

He reviewed the available data with reference to the new standard. Among the several reasons for selecting W K_{α_1} as a reference line, Bearden notes that this line is highly symmetrical and that its energy is large enough to be used in transmission-type crystal spectrometers and to calibrate γ-rays directly.

According to Bearden, the conversion factor

$$\Lambda^* = \lambda(\mathring{A})/\lambda(\mathring{A}^*) \tag{2}$$

should be equal to 1.000 000 ± 5 ppm. In their 1969 adjustment of the fundamental constants, Taylor, Parker and Langenberg [8] conclude that

$$\Lambda^* = 1.000\ 0197\ \ (56) \tag{3}$$

or 20 ppm larger than unity (see Ref. [8], Table 35). They conclude that there is no particular advantage in using the new reference standard. We do not agree with this opinion, especially since Bearden's compilation is of much practical use. The revised energy of the W K_{α_1} X-ray is

$$59.319\ 18\ (\pm\ 6\ \text{ppm})\ \text{keV} \tag{4}$$

TABLE I. ADJUSTED VALUES OF m_0c^2

year	rest mass energy of electron m_0c^2 (keV)
1947	510.79 ± 0.06
1952	510.984 ± 0.016
1955	510.976 ± 0.007
1963	511.006 ± 0.005
1969	511.0041 ± 0.0016

TABLE II. ^{198}Au TRANSITION ENERGY

	1963 constants	1969 constants
Electron rest mass m_0c^2 (eV)	511 006±5	511 004.1±1.6
Correction for positronium binding energy (eV)	3	3
Uranium K_{α_1} X-ray energy ($K - L_{III}$) (eV)	98 442±3	98 441 ± 3
Experimental energy difference (511 K - 411 L_{III}) (eV)	766±6	766 ± 6
^{198}Au γ-ray energy (eV)	411 795±9	411 794 ± 7

2.2. The ^{198}Au 411 keV transition and m_0c^2

The most obvious way of defining the energy scale in nuclear spectroscopy is to relate it to the electron rest-mass energy m_0c^2. It turns out that there are some problems in doing this. First of all, the accepted value of the constant has been quite variable as is shown in Table I.

Secondly, as is well known, the annihilation peak is broadened by the Doppler effect, the centre of mass of the annihilating electron-positron pair being not at rest in the laboratory system; the line shape is then not the same as that of a normal γ-ray. In addition, the line is shifted, owing to the binding energy of the electron with which the positron annihilates. The peak, therefore, cannot be used as a standard without applying special care, as is demonstrated by Mauron et al. [9] (appendix A). The annihilation peak is normally shifted by several tens of eV. Therefore, most of the calibrations which rely directly on this line have to be rejected. This does not apply to the work performed by Murray, Graham and Geiger [10], who carefully compared the momentum of the ^{198}Au 411 keV L III external conversion line with the momentum of the K external conversion line of a sharp (not Doppler-broadened) component of the annihilation radiation produced in ice. The energy of the ^{198}Au 411 keV transition given in Table II has been deduced from this measurement. The new U K_{α_1} X-ray energy has been obtained from an adjustment of the value given by Bearden [7], using the value in Eq. (3) for Λ^*.

In view of this careful – but single – determination, many authors have used this ^{198}Au γ-ray as a reference standard.

2.3. Interrelation of the scales

It is, of course, most interesting to know how the energy scales are interrelated. By Knowles [11, 12] and by Van Assche et al. [58] the W K_{α_1} X-ray and the annihilation energies have been compared directly, using crystal spectrometry. Greenwood et al. [13] have compared the ^{183}Ta 406 keV and the ^{198}Au 411 keV lines with a Ge(Li) diode, the first transition

TABLE III. COMPARISON OF ENERGY SCALES

Reference	Relative increase necessary to adapt energies measured in the W K_{α_1} scale to the ^{198}Au or the m_0c^2 scale (ppm)	experimental method
Knowles [11] [8]	- 8 ± 38	$\lambda_A - \lambda(W\ K_{\alpha_1})$
Knowles [12] [8]	57 ± 15	$\lambda_A - \lambda(W\ K_{\alpha_1})$
Van Assche et al. [58]	14 ± 32	$\lambda_A - \lambda(W\ K_{\alpha_1})$
Greenwood et al. [13]	0 ± 28	$E_{411} - E_{406}$
Reidy and Wiedenbeck [14]	-18 ± 42	$\lambda_{411} - \lambda(W\ K_{\alpha_1})$
Piller et al.[15]	2.3 ± 11	$\lambda_{411} - \lambda(W\ K_{\alpha_1})$

was calibrated on the W K_{α_1} scale. Finally, Reidy and Wiedenbeck [14] and Piller, Beer and Kern [15] have measured the W K_{α_1} line in the ^{198}Au energy scale with curved crystal spectrometers. The results of these experiments are given in Table III.

The value from Ref. [12] was disregarded by Taylor et al. [8] in their adjustment since it appeared to be inconsistent with the remaining data. If it is excluded, we note that the two scales are quite consistent.

A one-ppm measurement of the Compton wavelength h/m_0c in a e^+- e^- annihilation experiment is planned by Sauder [52].

3. ERRORS

The meaning of the errors associated with the published values is often quite ambiguous. To clarify the situation, let us review the various errors involved in the determination of a γ-transition energy, with a crystal or magnetic spectrometer.

The first error, which will be called the measurement error F_1, is due, first, to the inaccuracy of the determination of the peak location on the scale (because of the statistical fluctuations of the measured points) and, secondly, to the non-linearity of the scale itself.

Then, the measurement error on the reference peak in a particular measurement will introduce a first scale error. The composition of F_1 with it gives the so-called relative error F_2.

Finally, the error on the absolute value of the reference standard energy introduces a further error in the scale. Its composition with F_2 gives the absolute error F_3. The analysis of errors is not so simple in calibrations with Ge(Li) detectors.

Sometimes, it occurs that an experimentalist determines the energies E_γ of a number of transitions belonging to one isotope for the same calibration of his apparatus. If the number of lines exceeds the number of levels

which they connect, an overdetermined set of linear equations has to be solved. A least-squares fit gives the best statistical estimates for the excitation energies and exitation energy differences – equal to the transition energies, after subtraction of the recoil energy – and for their errors (standard deviation). It is to be noted that the fit is insensitive to an error in the scale, because the energy sum combinations would just as well be satisfied if the energy scale were multiplied by some arbitrary factor close to unity: the procedure cannot detect errors in the energy scale unit. These statistically evaluated errors, which we shall simply call statistical errors F_1' normally are smaller than the corresponding measurement errors F_1, because of the averaging procedure. (The above remarks do not exactly apply to measurements performed with magnetic spectrometers since the observed momenta are not proportional to the transition energies.) The errors F_1' have to be combined with those associated with the position and energy of the reference peak to obtain new relative and absolute errors F_2' and F_3'. Whenever calibration lines are based on a single reference standard, as happens very often, the errors F_2 or F_2' are then relevant in the application of the Ritz combination principle. Small errors are especially necessary for the rejection of accidental energy combinations in the study of complex level schemes.

4. PRIMARY STANDARDS

As primary standards, we define transitions which have been precisely and directly calibrated with respect to one of the two reference standards, W K_{α_1} or ^{198}Au 411 keV. Only determinations with iron-free magnetic spectrometers or crystal spectrometers will be considered, these instruments being the most reliable.

4.1. ^{182}Ta

Low-energy transitions, up to 264 keV, have been measured with curved crystal spectrometers by Seppi et al. [16], Gruber et al. [17] and Piller and al. [15]. Unfortunately, indications on the calibration are missing for the results of the first of these investigations. The results of Gruber are calibrated against W K_{α_1} and those of Piller against both W K_{α_1} and ^{198}Au. They are, generally, in agreement. Table IV presents the results from Ref. [15].

4.2. ^{192}Ir

The seven most intensive lines in the decay of ^{192}Ir to ^{192}Pt have been calibrated by Muller et al. [18] and by Bergvall [19] with curved crystal spectrometers. The results of Muller have been updated by an average adjustment of the observed Os, Ir and Pt K_{α_1} and K_{α_2} wavelengths to the values tabulated by Bearden [7] and by the use of the constant

$$E\lambda(\mathring{A}^*) = 12.398\,301\,(\pm\,5.9\text{ ppm})\,[\text{MeV}\cdot\text{m}\,\mathring{A}^*] \qquad (5)$$

proposed by Taylor et al. [8]. The errors are those quoted by the authors. The data of Bergvall are calibrated against the W K_{α_1} line, for which an

TABLE IV. LEVEL-SCHEME-ADJUSTED TRANSITION ENERGIES FROM ^{182}Ta (^{198}Au 411.794 ± 0.007 keV SCALE). DATA OBTAINED WITH A CURVED CRYSTAL SPECTROMETER [15]

E_x (keV)	F_1' (eV)	F_2' (eV)	F_3' (eV)
31.7370	0.4	0.5	0.7
42.7143	0.9	1.0	1.2
65.7219	1.1	1.2	1.7
67.7496	0.2	0.6	1.3
84.6802	0.5	0.9	1.7
100.1067	0.3	0.9	1.9
113.6677	0.9	1.3	2.3
116.4172	0.6	1.1	2.3
152.4298	0.5	1.3	2.9
156.3819	1.0	1.6	3.1
179.3895	0.9	1.7	3.5
198.3478	0.8	1.8	3.7
222.1037	1.0	2.1	4.3
229.3162	3.0	3.5	5.2
264.0697	0.8	2.3	5.0

TABLE V. TRANSITION ENERGIES IN keV FROM THE ^{192}Ir DECAY

Muller[18]	Bergvall[19]	Murray[20]
295.950±0.031	295.961±0.021	295.937±0.009
308.461±0.033	308.445±0.022	308.428±0.010
316.469±0.034	316.510±0.020	316.485±0.010
467.995±0.061	468.088±0.050	468.053±0.014
588.41 ± 0.28	589.16 ± 0.23	588.557±0.017
604.54 ± 0.59	604.54 ± 0.24	604.385±0.017
612.88 ± 0.61	612.77 ± 0.25	612.435±0.017

TABLE VI. TRANSITIONS IN ^{60}Ni

Murray et al. [20] (iron-free magn. spectr.)	Reidy and Wiedenbeck [21] (crystal spectrometer)
1173.229 ± 0.040	1173.223 ± 0.080
1332.487 ± 0.046	1332.52 ± 0.100

energy of 59.319 47 keV was assumed. We have adjusted his results to the value in Eq. (4) reported in sub-section 2.1.

The same transitions have been observed in internal and external conversion by Murray et al. [20]. Their adjusted mean results are shown in Table V. They are considerably more precise than the values obtained with curved crystal spectrometers, especially for the transitions around 600 keV. An effective test of the combination principle can only be performed with the magnetic spectrometer data. Unpublished results by Reidy and Wiedenbeck are quoted in Ref. [14].

4.3. ^{60}Co

The two 1173 and 1332 keV transitions in ^{60}Ni have been measured by Murray et al. [20] and by Reidy and Wiedenbeck [21]. Their adjusted results are given in Table VI. They are in good agreement.

4.4. Other isotopes

The authors of Ref. [20] have further calibrated γ-rays in ^{24}Mg and ^{208}Pb, which can be used as primary standards. Many further lines have been observed with several different iron-free β-spectrometers and calibrated on the ^{198}Au scale. Reidy and Wiedenbeck [22, 23], for example, have also measured transitions in the decays of ^{24}Mg, ^{46}Sc, ^{56}Mn and ^{140}La, with a curved crystal spectrometer. All these results are of much poorer precision than those of Ref. [20].

A point of concern is that there is a gap between 612 and 1173 keV where we have no good primary standard of precision comparable to the data of Murray et al. [20].

5. METHODS OF OBTAINING SECONDARY STANDARDS. RESULTS FOR $E_\gamma < 2$ MeV

5.1. Remark on the consistency of standard sets

We shall call secondary standards γ-ray transitions which have been calibrated with the help of the previously listed primary standards, mainly using Ge(Li) detectors. After a preliminary remark, we shall review some of the most significant results.

A set of standards useful for spectroscopists should mainly have the virtue of fulfilling true energy combinations of gamma rays. It is, in general, not so important if all standards are affected by the same relative systematic error, and for this reason we will call a set with the above properties a consistent set.

Primary standards may be affected by undetected systematic errors, and it may be questionable to take averages over values obtained in different laboratories, especially if this average cannot be made for all the transitions in the entire energy range of interest. Table V shows that the values obtained for ^{192}Ir by magnetic and crystal spectrometers are not in very good agreement. A systematic discrepancy seems to be further substantiated by the unpublished results of Reidy and Wiedenbeck reported in Ref. [14]. Most authors have taken the energies determined in Chalk River as primary standards, without averaging with other data. We shall follow this practice.

5.2. Electronic determination of the system non-linearity

Several electronic methods have been suggested to determine the non-linearity of a Fe(Li) spectrometer. Black and Heath [24] have used a precision mercury-switch pulse generator and obtained a precision of 50 to 70 eV for transitions between 145 and 1836 keV.

The method has been improved by Strauss et al. [25]. The differential response of the multichannel analyser is measured with a precisely linear sliding-pulse generator. A precision of 40 to 110 eV is quoted for γ-rays between 280 and 2760 keV.

More recently, McKee et al. [26] have calibrated their system by measuring the system's differential response with a mercury relay pulse generator whose amplitude is distributed over the range of the spectrum by a computer-controlled 15-bit digital-to-analogue converter (DAC).

Some of the problems associated with this type of method are: the electronic devices have to satisfy their specifications; the intrinsic non-linearity of the detector is not tested; the electronic and detector pulses may not have identical shapes. A further shortcoming of the method will be discussed in the next sub-section.

5.3. "Bootstrapping" with full-energy and double-escape peaks (pair-peak method)

If it is assumed that the energy difference between a full-energy and a double-escape peak is exactly $2m_0c^2$, it is then obvious that high-energy transitions can be calibrated by comparing, in a simultaneous measurement, the double-escape peak with precise standards with energies which are about 1 MeV smaller.

Potential sources of errors in this method have been discussed by Gunnink et al. [27]. The most important is due to the fact that the photoelectrons and the electron-positron pair emitted in the detector do not have an isotropic distribution. Depending on their direction of emission, the electrons are accelerated or decelerated in the detector electric field, thereby changing the number of ion pairs produced. This effect cancels to some extent for the electron-positron pairs, so that shifts between double-escape and full-energy peaks are shown to occur if the irradiation is not perpendicular to the electric field.

This has another very important consequence: when a transition is calibrated with a Fe(Li) diode, using standard procedures, all γ-rays have to reach the detector from the same direction; otherwise important shifts will occur, as is demonstrated, e.g. in Ref. [9], Fig. 10. It is also clear that non-linearities due to the acceleration or deceleration of charge carriers in the detector cannot be taken into account by an electronic calibration.

A nice application of the method has been demonstrated by White et al. [28], who calibrated the high-energy transitions of ^{182}Ta against the well-known low-energy γ-rays. Since the double-escape peaks are relatively weak, in this case, compared with the neighbouring full-energy peaks, a pair spectrometer was used: the pair and full-energy spectra were simultaneously analysed and stored in separate halves of the 1024 channel memory. Since the same ADC is used for the two resulting spectra and the address shift is only digital, the same non-linear calibration curve is applicable. Peaks which would otherwise overlap can be calibrated especially well. In this way, six transitions with energies between 1189 and 1289 keV have been calibrated with an accuracy of 25 to 70 eV. It must be noted that with this method a small part of the energy ($\sim$250 keV) is related to the W K_{α_1} scale and a larger part to the $m_0 c^2$ energy scale.

The same authors have calibrated the other lines appearing in the decay of ^{182}Ta, using standard techniques. Then they made a least-squares fit to obtain the best statistical estimates for the excitation energies. According to our discussion in section 3, the errors in these quantities are of type F_1'. Because the measurement of many calibration lines is involved in this work, F_2' should be only slightly larger than F_1'. For the high-energy lines, the statistical errors are 13 to 15 eV. This further suggests the good agreement between the different energy definitions.

5.4. "Cascade-cross over" and combined methods

Another useful method consists in comparing the sum of the energies of cascade transitions with that of the cross-over. This procedure should be safer than the pair-peak method. To be useful, the following conditions should be met: each of the cascade transitions should have a sufficient energy, so that an effective step forward can be obtained; the heights of all the peaks have to be comparable. A generalization of the method consists in selecting an isotope where a number of well-spaced levels are connected by a larger number of transitions satisfying the above conditions. An important advantage of the procedure is that the set of secondary standards which is obtained has precisely this property of consistency which we require (see sub-section 5.1).

In the energy range under consideration here, the decay of ^{110m}Ag is very suitable for the purpose. Combining the pair-peak method and this method, Kern [29] has calibrated the transitions following the ^{110m}Ag decay (see Table VIIa) together with a few other gamma rays from other isotopes. Because of the consistency requirement, primary standards determined by the Chalk River group have been used nearly exclusively; thus, in particular, no average values have been taken for the ^{192}Ir standards; the data are based on the 1963 value of the ^{198}Au 411 keV transition. As in the case of ^{182}Ta discussed in the preceding sub-section, the errors calculated in the least-squares fit of the transitions to the excitation energies are of the type $F_1' \sim F_2'$. The author has added to these errors a 4 ppm component, to take

TABLE VIIa. LEVEL-SCHEME-ADJUSTED ENERGIES [29] AND AVERAGE INTENSITIES [38, 41, 56] OF THE TRANSITIONS FROM THE ^{110m}Ag DECAY ($\tau_{\frac{1}{2}}$ = 253 d)

E_γ(keV)	F_1' (eV)	F_2' † (eV)	F_3' (eV)	I_γ (rel.)	ΔI_γ
446.790	15	16	18	3.6	0.2
620.305	13	15	18	2.9	0.1
657.720	7	10	16	100.0	-
677.590	13	15	20	12.0	0.4
686.965	13	15	20	7.3	0.4
706.650	14	16	21	17.3	0.5
744.245	13	15	21	4.65	0.2
763.930	14	16	22	23.8	0.6
817.995	11	14	23	7.7	0.3
884.655	11	14	22	78.9	1.5
937.445	14	17	25	36.2	.8
1384.230	14	20	32	27.6	.6
1475.710	9	17	34	4.47	.15
1504.955	11	20	35	14.7	.3
1562.235	12	20	36	1.31	.06

† A 10 ppm scale error has been estimated.

into account the uncertainty in the value of m_0c^2, with the implicit argument that the error in the ^{198}Au 411 keV to m_0c^2 comparison had to be disregarded, since the observed energy difference is not changed when a new adjustment of m_0c^2 is performed. The quoted errors can still practically be regarded as F_2' type errors, maybe with larger confidence. To conform with the principles of section 3, F_2' erros have been recomputed in assuming a 10 ppm scale error, and reported in Table VII a. Recently, Helmer et al. [42] have extended their work [13] on gamma-ray standards to energies up to 1300 keV. They use two groups of isotopes to extend the calibrated range. In the first group, none of the isotopes satisfies the conditions stated at the beginning of this sub-section. In the second group, only one loop in ^{160}Tb does. This is maybe one of the reasons why the two sets of standards that they obtain show a non-negligible discrepancy of 17 ppm above 800 keV. The adopted procedure has not provided an adequate check of the consistency of the results.

TABLE VIIb. ENERGY STANDARDS ON THE "CHALK RIVER ^{198}Au 411 keV" SCALE (EXCEPT FOR TRANSITION BELOW 280 keV)

Parent isotope	E_γ (keV)	ΔE_γ (eV)	ref.	Year	Comment
^{22}Na	1274.52	70	24	1967	
	1274.55	40	25	1969	
	1274.54	40			adopted
^{24}Na	1368.524	40	20	1965	prim. std.
	1368.58	110	25	1969	
	1368.53	40			adopted
	2753.916	120	20	1965	prim. std.
	2754.04	110	25	1969	
	2753.96	100			adopted
^{46}Sc	889.25	70	24	1967	
	1120.50	70	24	1967	
^{51}Cr	320.07	50	24	1967	
	320.11	40	25	1969	
	320.09	40			adopted
^{54}Mn	834.84	70	24	1967	
	834.81	40	25	1969	
^{57}Co	122.061	10	13	1970	
	136.471	10	13	1970	
	136.473	15	26	1971	
^{60}Co	1173.229	40	20	1965	prim. std.
	1173.23	40	34	1968	PP
	1173.22	40	25	1969	
	1173.23	40			adopted
	1332.487	46	20	1965	prim. std.

TABLE VIIb. (cont.)

Parent isotope	E_γ (keV)	ΔE_γ (eV)	ref.	Year	Comment
^{60}Co	1332.54	40	34	1968	PP
	1332.49	40	25	1969	
	1332.50	40			adopted
^{88}Y	898.010	70	24	1967	
	897.990	40	25	1969	
	898.010	30	29	1970	
	898.000	30			adopted
	1836.080	70	24	1967	
	1836.127	50	27	1968	PP
	1836.030	110	25	1969	
	1836.030	30	29	1970	PP
	1836.050	30			adopted
^{94}Nb	702.630	30	57	1971	
	871.104	35	57	1971	
^{140}La	328.752	30	27	1968	
	328.745	15	29	1970	
	487.032	30	27	1968	
	486.995	30	29	1970	
	487.010	30			adopted
	815.784	45	27	1968	sum 328+487
	815.735	40	29	1970	
	815.740	35	29	1970	sum 328+487
	815.755	30			adopted
	1596.200	40	27	1968	PP
	1596.170	25	29	1970	PP
^{203}Hg	279.185	10	60,13	1964	
	279.210	40	25	1969	

TABLE VIIb. (cont.)

Parent isotope	E_γ (keV)	ΔE_γ (eV)	ref.	Year	Comment
^{207}Bi	569.700	40	25	1969	
	569.653	20	29	1970	
	569.670	20			adopted
	1063.630	70	24	1967	
	1063.590	40	25	1969	
	1063.630	30	29	1970	
	1063.620				adopted
	1770.220	40	29	1970	PP
^{208}Tl(Th C")	583.139	23	20	1965	prim. std.
	583.190	110	25	1969	
	2614.475	100	20	1965	prim. std.
	2614.53	110	25	1969	
	2614.49	100			adopted
^{241}Am	26.348	10	61	1966	(uncorrected)
	26.345	1	62	1970	
	59.543	15	61	1966	(uncorrected)
	59.537	1	59	1968	

5.5. Review of some frequently used secondary standards

We shall survey in Table VII data published in recent years, which have been proposed as standards. We perform a selection according to the following principle: only data obtained with the "Chalk River ^{198}Au 411 keV" primary set of standards will be considered, because this set is of proven consistency and there is no other complete set of the same quality. Data whose accuracy can be checked by energy combinations are preferred. Unfortunately, for many widely used calibrations, such a test cannot be applied. Also some transitions in the upper range have been obtained only with precision by the pair-peak method, and are therefore on a mixed scale; they are noted by PP. No correction has been applied for the difference between the 1963 and 1969 value of the ^{198}Au 411 keV transition, except for primary standards.

Since all data are on the same scale, errors of type F_2 would be adequate for comparisons. Though most quoted errors are absolute (F_3), it has not been judged very prudent to manipulate them for a tentative reduction to relative errors (F_2).

6. HIGHER-ENERGY SECONDARY STANDARDS ($E_\gamma > 2$ MeV)

6.1. Radioactive standards

The methods sketched in the preceding section are, of course, applicable in this higher-energy region. Not many isotopes of sufficient practical half-life emit intense γ-rays in this energy range. Marion [5] suggested the use of ^{66}Ga and ^{56}Co. The first isotope has a half-life of only 9.4 hours, but its spectrum extends up to 4.8 MeV. It has been studied recently by Camp and Meredith [43] and by Phelps et al. [48]. The transitions from ^{56}Co have been studied with considerable precision by Barker and Connor [32], Gunnink et al. [27], Gehrke et al. [33], Phelps et al. [48] and Camp and Meredith [43]. The results are in good agreement; checks on the consistency show that it is good. Statistical best estimates for the transition energies, set equal to the excitation energy differences less recoil energy, calculated with the author's program LEVELFIT, are given in Table VIII.

6.2. Radiations from nuclear reactions

A number of precise standards have been determined in transitions following thermal-neutron capture. Marion [5] has reviewed results from

TABLE VIII. CONSISTENT ENERGIES AND AVERAGE INTENSITIES FOR THE MOST PROMINENT TRANSITIONS IN THE ^{56}Co DECAY. THE ENERGIES HAVE BEEN OBTAINED BY A LEAST-SQUARES FIT OF THE TRANSITIONS TO THE LEVEL ENERGIES, USING THE DATA OF Refs [27, 32, 33, 43, 48]. THE F_1' STANDARD DEVIATIONS HAVE BEEN COMBINED WITH AN ESTIMATED 10 ppm SCALE ERROR, TO OBTAIN THE F_2' ERRORS. THE INTENSITIES WERE OBTAINED BY AVERAGING DATA FROM Refs [32, 33, 38, 43, 48, 51]

E_γ (keV)	F_1' (eV)	F_2' (eV)	I_γ	ΔI_γ	E_γ (keV)	F_1' (eV)	F_2' (eV)	I_γ	ΔI_γ
787.79	60	60	3.3	0.3	2034.92	60	65	77.7	2.
846.74	25	25	1000.0	–	2113.36	75	80	3.8	0.1
977.46	60	60	14.5	0.7	2598.52	50	55	171.2	2.
1037.86	60	60	133.4	2.5	3009.80	55	65	9.4	0.5
1175.15	75	75	21.2	1.2	3202.20	60	70	31.5	0.5
1238.31	45	45	686.	4.	3253.56	50	60	75.9	0.9
1360.24	55	55	42.7	0.4	3273.18	55	65	16.9	0.4
1771.53	60	65	157.2	2.	3451.24	65	75	8.8	0.2
1963.93	65	70	7.0	0.3	3548.21	260	260	1.8	0.1
2015.30	55	60	29.8	0.5					

TABLE IX. LEVEL-SCHEME-ADJUSTED ENERGIES [34] AND INTENSITIES [53] OF THE MOST INTENSIVE TRANSITIONS FROM $^{53}Cr(n, \gamma)^{54}Cr$

E_γ (keV)	F_3	I_γ (rel)	ΔI_γ
834.87	.05	100.	
1784.67	.10	11.9	0.7
2239.07	.10	12.8	0.7
3719.73	.25	4.0	0.3
4847.01	.32	1.9	0.2
4872.27	.33	1.0	0.2
5999.51	.50	5.5	0.6
6645.31	.65	12.8	1.3
7099.66	.66	10.5	1.1
8884.08	.65	64.	7.

TABLE Xa. ENERGIES [37] AND INTENSITIES [37, 39, 40, 53, 54] OF THE γ-RAYS EMITTED IN THE DECAY OF ^{180m}Hf. THE ENERGIES DETERMINED BY REIERSON [54], WITH A CURVED CRYSTAL SPECTROMETER CALIBRATED WITH THE ^{198}Au 411 keV TRANSITION, ARE SYSTEMATICALLY LARGER THAN THE RESULTS OF GEINOZ [37], CALIBRATED ON THE CHALK RIVER SCALE. WE HAVE THE SAME SITUATION AS IN ^{192}Ir

E_γ (keV)	F_3	I_γ (rel)	ΔI_γ
57.545	.040	51.3	2.5
93.315	.020	18.2	.4
215.235	.015	86.1	.8
332.260	.020	100.	-
443.145	.025	88.5	1.3
500.690	.030	14.5	1.2

TABLE Xb. ENERGIES AND RELATIVE INTENSITIES [9, 53] OF X- AND γ-TRANSITIONS FOR A DETECTOR EFFICIENCY CALIBRATION. THE LINES IN ^{49}Ti AND ^{53}Co ARE OBSERVED IN ^{48}Ti(n, γ) AND ^{52}Co(n, γ) EXPERIMENTS

Isotope	Eγ (keV)	Iγ (rel)	ΔIγ %	Isotope	Eγ (keV)	Iγ (rel)	ΔIγ %
^{137}Cs	Kα	8.09	0.5	^{24}Na	1368	100.	-
	662	100.	-		2574	99.9	<0.2
^{198}Au	Kα	2.92	2.0	^{46}Sc	889	100.	-
	411	100.	-		1120	100.0035	$1.5 \cdot 10^{-3}$
^{57}Co	14	11.2	3.0	^{60}Co	1173	100.	-
	122	100.	-		1332	100.13	0.05
	136	12.05	1.3	^{228}Th	583	100.	-
					2614	117.4	1.
^{108m}Ag	432	100.	-				
				^{49}Ti	1497	100.	
	618	100.6	<0.2		4876	100.	
	727	100.7	<0.2				
				^{53}Cr	2319	100.	
^{22}Na	511	100.			5610	100.	
	1274	55.6	1.0				

currently used reactions. Recently, White et al. [34] have studied the ^{53}Cr(n, γ)^{54}Cr reaction by a combined approach, using the pair-peak and the generalized cascade-cross-over technique. The consistency of the results has been tested. The results are given in Table IX.

A particular problem regarding gamma-rays emitted after a charged-particle reaction arises from the fact that the radiating nucleus may not be at rest at the time of emission, causing a Doppler shift of the radiation. In addition, if the photon is emitted directly from a resonance, the width of the resonance, which is in general non-negligible, will broaden the observed peak. Marion [5] has also reviewed some useful reactions and energies.

7. INTENSITY CALIBRATIONS

7.1. Experimental methods

Intensity standards have been obtained by three methods.

The first method takes advantage of the property that intensity ratios can be deduced from the structure of the decay scheme. In general, corrections for electron conversion must be applied, but the present status

TABLE XI. PROTON-CAPTURE REACTIONS SUITABLE FOR INTENSITY CALIBRATIONS [44]. THE INTENSITIES OF THE CASCADES TRANSITIONS ARE EQUAL. GAMMAS EMITTED HAVE GENERALLY A NON-ISOTROPIC ANGULAR DISTRIBUTION. AT 55°, P_2 (cosθ) = 0 AND THE CORRECTIONS ARE SMALL FOR THE CASCADE UNDER CONSIDERATION. AT OTHER ANGLES CORRELATION COEFFICIENTS, AS GIVEN IN REF. [44], HAVE TO BE USED

Reaction	Proton energy (keV)	Cascade (Level energies in MeV)	Yield %
$^{27}Al(p,\gamma)^{28}Si$	2489	13.987→4.617→1.780→0	100
$^{23}Na(p,\gamma)^{24}Mg$	1318	12.957→1.369→0	90
$^{23}Na(p,\gamma)^{24}Mg$	1417	13.052→4.123→1.369	95
$^{23}Na(p,\gamma)^{24}Mg$	1394	13.030→5.235→1.369	55
$^{39}K\ (p,\gamma)^{40}Ca$	1344	9.643→3.905→0	50

of the theory allows exact results to be obtained even if the correction is large. Low-energy calibrations can use calculable X-ray to γ-ray intensity ratios [9, 49].

The second method consists in using calibrated isotopes, which are commercially available.

In the third method, the efficiency of the detector is calculated, see, e.g. Ref. [35], or a comparison is made with transitions calibrated with NaI(Tl) scintillators, whose efficiency has been calculated, as seen in Ref. [36].

7.2. Results

It is particularly interesting to use the same isotope for both energy and intensity calibrations. The transitions from ^{180m}Hf have been investigated in several papers [37, 39, 40, 53, 54, 55]. Results concerning this isotope and data on other suitable calibration isotopes [9, 53] are given in Table X. The intensities of the γ-rays from the decay of ^{110m}Ag have been calibrated by Aubin et al. [38], Brahmavar et al. [56] and Moragues et al. [41]. Average results are given in Table VIIa. Many papers [32, 33, 38, 43, 44, 48, 51] are devoted to the measurement of relative intensities in ^{56}Co (see Table VIII). At higher energies, the γ-rays in the decay of ^{66}Ga [5, 43, 48], those appearing in the reaction $^{14}N(n,\gamma)$ – see Ref. [5] – and γ-branching ratios from proton capture reactions [44] can be used for intensity calibration up to 12 MeV. The latter results are given in Table XI.

8. CONCLUSION

It has been shown that the available energy scales based on W K_{α_1}, ^{198}Au 411 keV and m_0c^2 are compatible within the present accuracy of the order of 10 ppm. There seems, however, to exist a discrepancy between

the primary standards obtained at Chalk River and with crystal spectrometers. Because of their high quality and because their consistency has been tested several times, the former set has been used preferentially by most experimentalists and, consequently, the standards tied to the "Chalk River ^{198}Au 411 keV scale" are the most reliable.

This situation is likely to change in the next few years because the precision obtainable with crystal spectrometers is considerably improving. This is for three reasons. Crystals with a few seconds mosaic spread and acceptable reflectivity in higher orders of reflexion are being investigated. Much work is performed to improve the precision of angle measurement with optical angle interferometers [45, 46, 50, 52]. The precision aimed at is a few milliarcseconds. Finally, work is in progress in several laboratories to determine the lattice constant of high-quality crystals by X-ray interferometry. A precision superior to 1 ppm is anticipated [47].

To gain a maximal profit of these developments, a new reference standard will be needed. The W K_{α_1} X-ray standard is not ideal in nuclear spectroscopy because it gives broad peaks in higher order of reflection, and, therefore, the statistical error in the position is relatively large. The energy of the gold standard is, on the other hand, quite large for crystal spectrometry. The Bragg angle is small even in higher orders of reflection so that the relative precision in the scale calibration is not so good as it could be. A low-energy transition in the ^{182}Ta decay, e.g. the 67.749 keV line, would be much more favourable. Since intense high-energy gamma-rays are present in the ^{182}Ta spectrum, causing shielding problems, we would also suggest the 84 keV transition in the decay of ^{170}Tm, a 130 d radioisotope, which can conveniently be produced by neutron activation (130 b) and which does not emit other gamma-rays.

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DISCUSSION

P.M. ENDT: You have extended your survey to high energies by means of (n, γ) transitions in ^{53}Cr, I think. What is the abundance of ^{53}Cr?

J. KERN: It is about 10%. But because of a favourable cross-section, the reaction with this isotope dominates.

P.M. ENDT: I want to point out, that van der Leun (Phys. Lett. 30B (1969) 406) has proposed the use of the (n, γ) reaction to calibrate (p, γ) work, superimposing both spectra. This can be done easily: you need a neutron source, e.g. a ^{241}Am-Be source. The Ge detector is surrounded by a shield, which will capture the neutrons and provide you with the capture gamma-rays that you use as calibration. Now, Cr would not be the most

suitable material. Van der Leun has used Fe; it is very easy to surround the detector with a Fe-cap. You put the neutron source into a water vessel, for neutron thermalization. An Am source costs something like $1000.-, it is relatively cheap. You need, of course, good (n, γ) standards. With Fe, you get a good enough statistics for the transitions leading to the ground state doublet in ^{57}Fe in something like 10 min. What we need, are a few isotopes, having a good cross-section, preferably a simple spectrum, not too many lines, otherwise it spoils the (p, γ) spectrum – a few lines well spread up to 10 MeV, if possible. Then all (p, γ) spectra could be expressed in terms of just a few reference lines, which should be known with good precision; 100 eV would be very nice. This is not yet available, but somebody should go into this and really do it.

C. ROLFS: In Table XI you give a few examples of proton-capture reactions suitable for intensity calibrations, and you give the yield of the transitions, at a ratio of 1:1:1, e.g. in the case of ^{27}Al. Do you have an idea of the error on this? As we have discussed previously, all these decays can go through many intermediate states. To obtain a good efficiency curve, say better than 5%, you have to do a very careful job to find all the other cascade members. I would almost say that one should go to much lighter nuclei, where the level density is lower, and where you can estimate these other contributions to the lower member in a better way.

J. KERN: Table XI is extracted from a work by Singh and Evans (Ref. [44] of my paper). The authors have searched for the presence of weak lines populating and de-populating the intermediate level (s) in the cascade. They have failed to identify such lines and have assumed that the error on the relative intensity of the gamma-rays in the cascade is ± 2%. I am sceptical about this precision, which I therefore did not quote in the table caption. In one month I was only able to make a survey and not to produce calibration lines up to 25 MeV with all the required accuracy.

C. ROLFS: In a nucleus with a high level density you really have the problem of having, say, 50 transitions to estimate, and of summing up all the upper limits, which may turn out to be quite large, so that you probably have to run for a month to get these limits down.

P.M. ENDT: I agree that this is dangerous.

C. ROLFS: One likes to get the intensity calibration together with the main measurement, and not make the calibration afterwards, when the situation may not be exactly the same.

P.M. ENDT: This brings us back to what had been said in a previous discussion – that generally the intensities of the lines which you see going to the ground state add up to a value which is larger than the sum of all primary lines in the spectrum because you probably lose weak primaries going to higher intermediate states.

S.S. HANNA: In analysing spectra taken with a Ge detector, do you have any comments, advice or recommendations as to how one should extract the position of a gamma-ray line?

J. KERN: It is first most important to use the same procedure for the analysis of the new lines and of the standards, so that any systematic error is the same for both sets of lines. Very often the peaks lie on a curvy background; the lines are not exactly symmetrical, they have tails in their bottom part, toward their low-energy side. The analysis of the lower part of the lines is, therefore, not always very easy to perform. For this reason, some people fit only the top of the line, which is rather

well symmetrical. I do not stick to this method, because you first lose quite a bit of information, secondly, you make errors whenever the assumptions on the background slope are bad and when you have close multiplets, especially with appreciable intensity ratios. It is better to fit the line shape as closely as possible.

S.S. HANNA: When you do that, do you calculate the centroid, do you fit with a Gaussian?

J. KERN: I said before that if you make an error, it ought to be systematic, and apply equally to all peaks. This is difficult in practice, because you have not the same statistics for each peak; you have got to best reproduce analytically the true shape of the lines. Only the top, and generally the high energy edge of the line is Gaussian. The low-energy side can be fitted according to a procedure described in Ref. [29]: you first add a constant component to the background proportional to the peak height. Then you deform your line profile in adding components which you obtain in multiplying the Gaussian with a simple polynomial, having terms of degree 4 and 12. This already gives, in general, a quite good fit. If this is not yet sufficient, you can add still a further, so-called, "exponential tail", with a generally longer range and adjustable amplitude. The shape parameters, maximum 5, have to be obtained by fits of peaks with good statistics. Average or smoothly varying values have then to be used in a systematic way over the entire spectral range. One obtains very good fits. In the decay of ^{165}Dy, e.g., I was able to very well fit small lines on the low-energy side of a huge transition (see Ref. [29]). When you have poorer statistics, a pure Gaussian may be good enough. The procedure has been programmed. In the output you obtain the centroid of the Gaussian component, its area and the integrated area of the deformed peak.

M.A. MEYER: I think it is not generally true that the background is higher on the low-energy side of the double escape peak of high-energy gamma-rays.

P.M. ENDT: You can get some sort of a hole on the low-energy side first, and then a rise in the background.

J. KERN: This can happen, but is not significant in high-energy (n, γ) capture work. The analytical shape could be easily modified to take into account this feature, if necessary.

P.M. ENDT: Could you comment on the annihilation peak, why is it broader and shifted?

J. KERN: The broadening is easy to understand. The electron-positron annihilating pair is normally not at rest and has a cinetic energy of a few eV. The photons have to take over the corresponding small impulse of the centre of mass. To do this, and because the photons are so much lighter than the electrons, their energies are changed by quantities of the order of keV. This can easily be calculated in using the linear momentum conservation law or in calculating the Doppler effect due to the speed of the annihilating pair. As a further consequence, the two quanta are not emitted with a relative angle equal to exactly 180°.

The reason for the shift is more complicated. In the case of positronium annihilation, you have a 3.4 eV shift, half the binding energy. You have, in general, other shifts, which are material-dependent and are primarily due to the binding energy of the pair in the surrounding material. These shifts may amount to some tens of eV (see, e.g. Ref. [9] of my paper, Appendix A).

The narrow component observed in ice (see Ref. [10] of my paper) is not even unambiguous, as the annihilation in ice is not fully understood. That line is not necessarily produced exclusively by positronium annihilation and may also be shifted by a few eV. A fine experiment which somebody ought to do is to measure annihilation in some material where the process is well understood, like potassium, and properly unfold the result.

P.M. ENDT: I see, so your advice would be not to use the annihilation line, just never.

J. KERN: This is exactly what I mean.

M.A. MEYER: You touched the problem of bootstrapping with the full and double escape peaks. How reliable is this?

J. KERN: It depends on the detector and on the geometry of irradiation. The problem is discussed in Ref. [27] of my paper. In addition, some diodes show shifts depending on the source-detector distance. Still other problems arise if the time structure of the calibration peaks is not the same as that of the observed spectrum. To come back to your question, the energy difference is equal to 2 m_0c^2 to a precision of probably 15-20 eV, if the experiment is done properly. This is what I infer from the results of Ref. [28] of my paper.

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