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The precision determination of some γ-ray energies using the Fribourg curved crystal spectrometer

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Abstract The energies of several γ-rays from the decay of ^{169}Yb, ^{170}Tm and ^{192}Ir have been determined with the Fribourg curved crystal spectrometer, using a 4 mm thick [110] quartz plate. The most intense lines have been observed with reflection orders up to n = 10 and 12. Relative precisions down to a few ppm have thus been obtained. The spectrometer was calibrated with the 411.794 ± 0.007 keV γ-ray transition in the decay of ^{198}Au.

1. Introduction

With presently available semiconductor detectors, it is possible to determine the energy of gamma rays with a precision of a few electron-volts, provided sufficiently accurate energy standards have been determined in the energy range of interest. This condition is fulfilled below 280 keV; at higher energies the situation is not so satisfactory. In the energy range of 280 to 620 keV, the γ-ray transitions following the decay of ^{192}Ir to ^{192}Pt have been most frequently used for calibration purposes. These lines have been calibrated against the 411 keV transition from the decay of ^{198}Au by Murray et al.[1], who used an iron-free β-spectrometer, and against the tungsten K_{α_1} X-rays by Bergvall[2], with the help of a curved crystal spectrometer. In order to compare these results, the scale inter-relation has to be known. Kern[4] has adjusted the curved crystal spectrometer data to a scale where the W K_{α_1} X-ray energy is 59.31918 keV, as recommended by Taylor et al.[5], and the β-spectrometer data to a scale where the ^{198}Au γ-ray energy is 411.794 ± 0.007 keV. The latter value results from a reevaluation of the original external conversion experiment of Murray et al.[3], who used an Uranium radiator. In the computation, the constants proposed by Taylor et al.[5] and the U K_{α_1} X-ray energy adjusted to the above X-ray energy scale, had been used. The two sets of results on ^{192}Ir were found not to be in good agreement. Recently Beer and Kern[6] determined the W K_{α_1} X-ray energy to be 59.31737 ± 0.000 54 keV in a scale where the ^{198}Au γ-ray has the value 411.794 ± 0.000 keV, a result valid if a ^{187}W radioactive source is used to produce the W X-rays, as was done in Bergvall's experiment. This new determination implies, in turn, a recomputation of the ^{198}Au γ-ray energy, since the U K_{α_1} X-ray energy has to be adjusted. The following results are obtained:

^{198}Au γ-rays	411.797	± 0.007	keV
W K_{α_1} X-rays	59.3178	± 0.0012	keV
U K_{α_1} X-rays	98.438		keV

It must be noted that the X-ray energies are very close to the values proposed by Bearden[7].

Using these values, the adjusted energies of Bergvall and of Murray et al. reported in Table I are obtained. The quoted errors are those given in the original papers. Inspection of Table I shows that, though the results are generally compatible within the errors and are in better agreement than in the previous comparison[4], the data obtained with the curved crystal spectrometer are still systematically larger than those of Murray et al.

Besides, the relatively large errors on the energy of the lines around 600 keV in the work of Bergvall do not allow a conclusive comparison between the cross-over energies and the energy sums of the corresponding cascade transitions. The aim of the experiments described in this paper is to improve the energy determination of the lines appearing in the ^{192}Ir to ^{192}Pt decay. Results on transitions appearing in the ^{169}Yb and ^{170}Tm decays are also reported (Table IV). The energies of transitions from the ^{182}Ta decay, as determined by Piller et al.[8], but with recomputed errors, are also given (Table V). All the data are given in the 411.794 keV ^{198}Au scale, for the following reasons: a) Many precision results on this scale have been published recently. The comparison is thus easier. b) The value 411.797 keV was obtained in using the careful but single measurement by Beer and Kern[6]. In that experiment the ^{187}W source was very deep (4 mm). A computer simulation indicates that the source geometry should not cause any sizeable shift. Nevertheless, the results should be confirmed by an independant measurement, by preference with a spectrometer in the Cauchois geometry. c) A scale adaptation for crystal spectrometer results is most easily done, since it is linear.

2. The experiments

The instrument used for the experiments is the Fribourg, DuMond type, curved crystal spectrometer, described in a previous paper[8]. Before the beginning of the measurements reported here, a modification had been made to the apparatus: The table supporting the crystal and the crystal driving mechanism have been mechanically isolated from the main frame of the spectrometer (which supports the collimator), and fastened directly to the ground through two concrete pillars. By this change, the influence from rotation of the heavy collimator on the crystal axis is practically eliminated, resulting in a smaller non-linearity of the angular scale. This has been recalibrated with the Moore table[9] and with an improved auto-collimator[10], resulting in an average scale precision of 0.15 arcsec.

The crystal, a 4 mm thick [110] quartz lamina, was bent to a 5 m radius of curvature by the imprisonment method[11]. The effective reflecting area, selected with a lead collimator, was approximately 5 cm^2. Using a 0.05 mm thick source, the reflexes had a width of 3 arcsec FWHM. During the whole experiment the spectrometer was enclosed in a thermostatized hut maintained at a temperature of 19.55 ± 0.05° C. Each measurement consisted in the observation of the reflexes on the right and on the left. Since the corresponding reflexes have the same shape, the errors due to the change in line shape with angle are suppressed. In the analysis of the data the line profile was fitted with a deformed gaussian[12], using a least squares procedure[13]. The radioactive sources were produced by neutron irradiation of natural metallic Au,Co,Ag, Ta, W and Ir strips,natural Tm oxide and to 18.25 % enriched ^{168}Yb oxide powder. There dimensions were generally 0.05 x 4 x 32 mm^3.
The measurements were made in the following time sequence:
Ir, Au, Ag, Yb, Tm, Co, W, Au, Ta.

Before the second ^{198}Au measurement, the crystal was slightly turned against its supporting table, in order to minimize the effect of systematic errors on the angular scale calibration. No time dependent effect was observed. The data obtained with the Ta and W sources are reported elsewhere[6]. Those obtained with the Ag and Co sources are not yet evaluated.

3. The results

The results of the experiments are presented in Table II. For each transition, the quantities $\sin\theta/n = \lambda/2d$, resulting from the measurement of the left to right angular differences observed in several orders of reflexion n, are given. Each value is followed by an error σ_i, obtained by the composition of the statistical uncertainties (standard deviations) on the line angular positions and of the estimated 0.15 arcsec error on the spectrometer scale calibration. The weighted average is followed by the internal (σ_I) and external (σ_E) errors. The first is the expected error computed from the a priori errors σ_i, while the second, given between parenthesis, is estimated from the deviations of the individual values from the mean. A comparison of these errors shows that in most cases σ_E is about equal or smaller than σ_I. This indicates that our estimated 0.15 arcsec error on the scale calibration is correct and not underestimated.

The spectrometer was calibrated with the ^{198}Au 411.794 ± 0.007 keV transition. The energies of ten transitions from the decay of ^{192}Ir to ^{192}Pt are reported in Table III, col. 1. In col. 2 are given the measurement errors ΔE_1 corresponding to the values σ_I in Table II. Since the transitions are placed between five levels in ^{192}Pt, the most probable level energies can be computed, using the least squares fit method. This was done with help of the computer program LEVELFIT, which takes into account the atomic recoil. The level energy differences, which are the best statistical estimates for the transition energies, are reported in col. 3, and followed with statistically estimated standard deviations $\Delta E_1'$ (col. 4). Both the ΔE_1 and $\Delta E_1'$ errors do not include the uncertainties due to the calibration. The composition of the statistical errors $\Delta E_1'$ with the 4 ppm error on the $\lambda/2d$ value for gold (Table IIa) gives the relative errors $\Delta E_2'$(Au), while the composition of the $\Delta E_2'$(Au) with the 17 ppm error on the ^{198}Au reference energy yields the absolute errors $\Delta E_3'$.

The energies of two further transitions, one from the decay of ^{169}Yb, the other from that of ^{170}Tm, are given in col. 2, Table IV. The measurement error ΔE_1, the relative error ΔE_2 and the absolute error ΔE_3 are given for each line in col. 3 to 5. These data are on the ^{198}Au 411.794 ± 0.007 keV scale.

4. Discussion

Inspection of Table III shows that the differences between the adjusted transition energies E_γ' and the observed values E_γ are always smaller than the estimated errors ΔE_1. The computed chi squared is then smaller than unity. This fact and the effective use of the system overdetermination have as a consequence smaller statistical errors $\Delta E_1'$. In some cases these are much smaller than the measurement errors ΔE_1. At the level of precision obtained here, no deviation from Bragg's law nor any secondary effect has been observed.

In nuclear spectroscopy, precision is generally needed to reduce the number of accidental combinations in using the Ritz combination principle. If all calibration lines are on the same ^{198}Au scale, the energy error on the reference standard is irrelevant and the relative errors $\Delta E_2'$ have to be used. A comparison of col. 4 and 5, Table III, shows that in spite of a careful measurement of the ^{198}Au 411 reference line, the $\Delta E_2'$ errors are in several cases much larger than the $\Delta E_1'$ errors. This is because the reference energy is fairly large for crystal spectroscopy, the Bragg angle is small, and a good relative precision is difficult to achieve. The use of the tungsten K_{α_1} X-ray cannot be recommended, since these X-rays have an important natural width and because their energy depends on isotopic composition[14), chemical environment[15) and production mode[6). A low energy γ-ray, i.e. the 63 keV transition from the ^{169}Yb decay (32 d) or the 84 keV line from the ^{170}Tm decay (127 d), would be more convenient reference standards. Both isotopes have reasonable life times and large neutron capture cross sections. If ^{168}Yb is used as a neutron target, high specific activities can be obtained. A special merit of ^{170}Tm is that no other γ-ray is emitted in the decay, so that the health hazards in the manipulation of this kind of radioactive source can be minimized. As an example, we give in Table III, col. 6, the relative precisions obtained with reference to a ^{169}Yb source. They are in many cases two times smaller that with the ^{198}Au standard (col. 5). Absolute determination of the wave length of the 63 and 84 keV γ-rays are planned at the U.S. National Bureau of Standards[16).

The present ^{192}Ir results, adjusted to the 411.797 keV ^{198}Au scale, are given in Table I. It is seen that they are generally intermediate between those of Murray <u>et al.</u>[1) and those of Bergvall[2). They agree with the results of some other recent determinations[17)18)19).

For the sake of easy reference, the results of Piller <u>et al.</u>[8) on ^{182}Ta are given in Table V. The errors have been recomputed.

As a final remark, it must be emphasized that all errors have the meaning of standard deviations. Higher confidence limits are often desirable for reference lines and a 2σ intervall may be chosen.

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Table captions

Table I — Transition energies in keV from the ^{192}Ir decay determined in this and in previous works. The data have been adjusted to a scale where the ^{198}Au γ-ray energy is 411.797 ± 0.007 keV (see text).

Table IIa — Measurement of the 411 keV line in the ^{198}Au decay. The internal (σ_I) and external (σ_E) errors on the mean are given. The second stands between parenthesis.

Table IIb — Measurement of lines in the ^{192}Ir decay. The internal (σ_I) and external (σ_E) errors on the mean are given. The second stands between parenthesis.

Table IIc — Measurement of γ-rays in the ^{169}Yb and ^{170}Tm decays. The internal (σ_I) and external (σ_E) errors on the mean are given. The second stands between parenthesis.

Table III — Gamma-ray energy values from the decay of ^{192}Ir to ^{192}Pt (^{198}Au 411.794 ± 0.007 keV scale).

Table IV — Gamma-ray energy values from the decay of ^{169}Yb and ^{170}Tm (^{198}Au 411.794 ± 0.007 keV scale).

Table V — Gamma-ray energy values from the decay of ^{182}Ta, taken from ref. 8 and adjusted to a scale where the ^{198}Au line has the energy 411.794 ± 0.007 keV.

Bergvall (2)		Murray et al. (1)		Present work	
295.956	± 0.021	295.937	± 0.009	295.9505	± 0.005
308.439	0.022	308.429	0.010	308.4480	0.005
316.504	0.020	316.486	0.010	316.5000	0.006
468.078	0.050	468.057	0.014	468.0580	0.008
589.14	0.23	588.565	0.017	588.5605	0.011
604.52	0.24	604.393	0.017	604.3986	0.011
612.75	0.25	612.443	0.017	612.4505	0.011

Table I

Order n	10^8x (λ/2d)	Order n	10^8x (λ/2d)
1	612 788 $\pm$ 56	7	612 793 $\pm$ 7.6
2	612 823 26	7	612 792 7.4
3	612 787 17	8	612 788 8.0
4	612 787 13	8	612 794 6.5
4	612 799 13	10	612 797 9.8
5	612 806 10	10	612 792 5.3
5	612 792 10		
		mean	612 793 $\pm$ 2.5 (1.4)

Table IIa

Order n	^{169}Yb 10^8x (λ/2d)	Order n	^{170}Tm 10^8x (λ/2d)
2	3 997 963 $\pm$ 26	1	2 995 151 $\pm$ 52
3	3 997 977 17	2	2 995 137 26
4	3 997 953 13	3	2 995 132 17
5	3 997 975 10	4	2 995 123 13
		5	2 995 130 11
		7	2 995 130 9
mean	3 997 968 $\pm$ 6.9(5.7)	mean	2 995 129 $\pm$ 5.7(1.8)

Table IIc

Table IIb

Order n	10^8 x (λ/2d)	Order n	10^8 x (λ/2d)
	136.3 keV		316.5 keV
1	1 850 803 ± 53	1	797 305 ± 52
2	878 26	2	310 26
3	861 17	3	305 17
4	853 14	5	279 10
5	844 11	7	312 7.5
		8	305 6.8
		9	293 6.5
		10	304 5.4
		12	304 5.2
mean	1 850 851 ± 7.2(5.8)	mean	797 302.0 ± 2.6(2.8)
	295.9 keV		416.5 keV
1	852 691 ± 52	2	605 969 ± 26
2	699 26	3	944 18
3	672 17	4	927 13
4	681 13	5	941 11
5	659 11		
7	663 8		
8	664 7		
9	660 7		
10	663 6		
mean	852 663.8 ± 3.0(2.3)	mean	605 939.5 ± 7.2(6.3)

Order n	10^8 x (λ/2d)	Order n	10^8 x (λ/2d)
	308.4 keV		468.0 keV
1	818 145 ± 52	1	539 122 ± 52
2	135 26	2	165 26
3	108 17	3	135 17
4	104 13	4	120 13
5	116 11	5	136 10
7	105 8	7	134 7.5
8	105 7	8	136 7.5
9	116 7	10	146 6.2
10	125 6		
mean	818 114.2 ± 3.0(3.2)	mean	539 138.3 ± 3.5(3.1)
	588.5 keV		612.4 keV
3	428 766 ± 18	4	412 019 ± 17
4	748 13	5	026 10
5	751 10	7	030 8.2
7	759 8.2		
mean	428 755.6 ± 5.5(3.3	mean	412 027 ± 6.0(2.7)
	604.4 keV		884.5 keV
4	417 519 ± 13	2	285.267 ± 26
5	505 10	3	292 18
7	520 7.8	4	305 17
		5	291 11
mean	417 515 ± 5.6(4.8)	mean	285 292 ± 7.8(5.4)

Table II b (cont.)

1	2	3	4	5	6	7
Eγ (keV)	ΔE_1 (eV)	Eγ' (keV)	$\Delta E_1'$ (eV)	$\Delta E_2'$(Au) (eV)	$\Delta E_2'$(Yb) (eV)	$\Delta E_3'$ (eV)
136.3398	0.5	136.3400	0.4	0.7	0.5	2.4
295.9486	1.0	295.9483	0.6	1.4	0.8	5.2
308.4467	1.1	308.4464	0.6	1.4	0.8	5.4
316.4982	1.0	316.4977	0.6	1.4	0.8	5.6
416.452	5.0	416.4501	3.1	3.5	3.2	7.9
468.052	3.0	468.0546	0.8	2.1	1.1	8.2
588.551	7.6	588.5562	2.4	3.4	2.6	10.6
604.396	8.	604.3942	0.8	2.6	1.3	10.6
612.446	9.	612.4460	0.8	2.6	1.3	10.6
884.513	24.	884.5037	2.6	4.4	3.0	15.6

Table III

Eγ = measured γ-ray energies

Eγ' = least squares adjusted transition energies

ΔE_1 = measurement error

$\Delta E_1'$ = statistical error

$\Delta E_2'$(Au) = relative error, Au-scale (statistical error compounded with gold measurement error)

$\Delta E_2'$(Yb) = relative error, Yb-scale (statistical error compounded with Yb measurement error)

$\Delta E_3'$ = absolute error (relative error on gold scale, compounded with error on the energy of the gold line)

1	2	3	4	5
Isotope	E_γ (keV)	ΔE_1 (eV)	ΔE_2 (eV)	ΔE_3 (eV)
Yb-169	63.11823	0.11	0.28	1.1
Tm-170	84.25168	0.16	0.38	1.5

Table IV

1	2	3	4	5	6
E_γ (keV)	ΔE_1 (eV)	E_γ' (keV)	$\Delta E_1'$ (eV)	$\Delta E_2'$(Au) (eV)	$\Delta E_3'$ (eV)
31.7371	0.4	31.7370	0.3	0.4	0.7
42.7144	0.7	42.7143	0.5	0.6	0.9
65.7219	0.2	65.7219	0.2	0.6	1.3
67.7496	0.2	67.7496	0.2	0.6	1.3
84.6808	0.7	84.6802	0.5	0.8	1.6
100.1067	0.3			0.9	1.9
113.6673	1.2	113.6677	0.6	1.1	2.2
116.4152	1.7	116.4172	0.6	1.1	2.3
152.4285	1.4	152.4298	0.5	1.3	2.9
156.3821	1.2	156.3819	0.7	1.4	3.0
179.3909	1.8	179.3895	0.6	1.6	3.4
198.3483	2.2	198.3478	0.7	1.7	3.8
222.1013	2.4	222.1037	0.7	1.9	4.2
229.3162	3.0			3.5	5.2
264.0711	5.2	264.0697	0.7	2.2	5.0

Table V